Chirality and Quantum Spin - a Critical Assessment

822. WE-Heraeus-Seminar

02 – 05 December 2024

at the Physikzentrum Bad Honnef, Germany

WILHELM UND ELSE
HERAEUS-STIFTUNG

Introduction

The Wilhelm und Else Heraeus-Stiftung is a private foundation that supports research and education in science with an emphasis on physics. It is recognized as Germany's most important private institution funding physics. Some of the activities of the foundation are carried out in close cooperation with the German Physical Society (Deutsche Physikalische Gesellschaft). For detailed information see https://www.we-heraeus-stiftung.de

Aims and scope of the 822. WE-Heraeus-Seminar:

For electrons moving in chiral molecules, one spin projection is more likely to be transported through the structure than the other, despite that the chiral molecule is often diamagnetic. This is the chiral induced spin selectivity (CISS) effect. It is a quantum phenomenon that is based on chirality, spin-orbit coupling, particle-particle interaction, and compositeness. However, exactly how these aspects are brought together and result in the CISS effect is yet an enigma.

This question, among several others, will be discussed in this seminar, with the aim to reach some clarification. Leading experts working in the field of the CISS effect as well as expert physicists, chemists, engineering and biology-oriented scientists aiming to focus research in this direction will provide lectures with a broad scope to facilitate a wider comprehension and open discussions of this enigmatic issue.

Invited speakers will outline the contemporary state of the experimental and theoretical sides in presentations in order to open for questions and discussions between the attending researchers. In addition, participants are asked and encouraged to take part in the poster session preceded by an oral brief presentation of the poster contents.

Scientific Organizers:

Prof. Dr. Jonas Fransson	University of Uppsala, Sweden e-mail: Jonas.Fransson@physics.uu.se
Prof. Dr. Carmen Herrmann	University of Hamburg, Germany e-mail: carmen.herrmann@uni-hamburg.de

Introduction

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Registration:

Mojca Peklaj (WE Heraeus Foundation)
at the Physikzentrum, Reception Office
Sunday (17:00 h - 21:00 h) and Monday morning

Program

Program

Sunday, 01 December 2024

17:00 – 21:00	Registration	
18:00	*BUFFET SUPPER and informal get together*	

Monday, 02 December 2024

07:30	*BREAKFAST*	
08:15 – 08:30	Scientific Organizers	**Welcome**
08:30 – 09:15	David Waldeck	**Experimental Investigations of Spin and Charge Transport in Chiral Materials**
09:15 – 10:00	Magalí Lingenfelder	**How to Use Chiral or Magnetic Fields to Boost Clean Energy Processes**
10:00 – 10:45	Alexander Kuhn	**Fine-Tuning ORR Activity at Chiral Imprinted Mesoporous Metal Surfaces**
10:45 – 11:30	*COFFEE BREAK*	
11:30 – 12:15	Mathieu Gonidec	**Functional Coordination Compounds and the CISS Effect**
12:15 – 13:00	Nicola Spaldin	**Have We Learned Anything from Studying Chiral Phonons that Might Be Relevant for CISS?**
13:00 – 13:15	**Conference Photo (in front of the lecture hall)**	
13:15	*LUNCH*	

Program

Monday, 02 December 2024

14:30 – 15:15	Angela Wittmann	**Chiral-induced Unidirectional Spin-to-charge Conversion**
15:15 – 16:00	Michael Thoss	**Influence of Electronic-vibrational Coupling on Spin-selectivity in Chiral Molecular Junctions**
16:00 – 16:45	Bart van Wees	**Mechanism for Electrostatically Generated Magnetoresistance in Chiral Systems without Spin-Dependent Transport**
16:45 – 17:30	*COFFEE BREAK*	
17:30 – 18:15	Jacopo Simoni	**Ab Initio Spin Dynamics and Decoherence in Solids**
18:15 – 19:00	Shinji Miwa	**Microscopic Origin of Magnetoresistance in Chiral Molecule/Ferromagnet Junctions**
19:00	*DINNER*	
20:30	**Poster Flash Presentations**	

Program

Tuesday, 03 December 2024

07:30	*BREAKFAST*	
08:30 – 09:15	Daniel Bürgler	**Enantioselective Adsorption and Spin-selective Electron Transport of Discrete Helicenes on Cobalt**
09:15 – 10:00	Katharina Franke	**Wave-function Engineering on Superconducting Substrates: Chiral Yu-Shiba-Rusinov Molecules**
10:00 – 10:45	Christoph Tegenkamp	**The Mechanism of the Molecular CISS Effect in Chiral Nanojunctions**
10:45 – 11:30	*COFFEE BREAK*	
11:30 – 12:15	Latha Venkataraman	**Electronic Transport Across Chiral Molecules**
12:15 – 13:00	Thomas Fay	**A Molecular Perspective on the CISS Effect**
13:00	*LUNCH*	
14:30 – 15:15	John Abendroth	**Magnetic Field Effects and Spin-Correlated Radical Pairs in Chiral Donor-Bridge-Acceptor Molecules**
15:15 – 15:35	Tatsuhiko Ohto	**First-principles Study on the Electron-vibration and Spin-phonon Couplings of Chiral Molecules**
15:35 – 15:55	Jakob Staab	**Ab Initio Modelling of Chirality Induced Spin Selectivity**
15:55 – 16:05	**Video WE Heraeus Foundation**	
16:05 – 16:30	*COFFEE BREAK*	
16:30 – 19:00	**Poster Session**	
19:00	*DINNER*	

Program

Wednesday, 04 December 2024

07:30	*BREAKFAST*	
08:30 – 09:15	Helmut Zacharias	**The Spin of Photoelectrons from Monolayers of Helical Molecules**
09:15 – 10:00	Yossi Paltiel	**Chiral Spintronics Using Chiral - Chiral Interactions**
10:00 – 10:45	Jaemin Lim	**Analyzing CISS Mechanisms through Spin-Dependent Dispersion Relations**
10:45 – 11:30	*COFFEE BREAK*	
11:30 – 12:15	Yonatan Dubi	**Surface Magnetic Stabilization and the Photo-emission Chiral-induced Spin-selectivity Effect**
12:15 – 13:00	Joseph Subotnik	**Momentum Conservation and Phase Space Approaches to Electronic Structure**
13:00	*LUNCH*	
14:30	**Excursion**	
18:30	*HERAEUS DINNER* *(social event with cold & warm buffet with complimentary drinks)*	

Program

Thursday, 05 December 2024

07:30	*BREAKFAST*	
08:30 – 09:15	Ron Naaman	**CISS - More Surprises to Come**
09:15 – 10:00	Jeanne Crassous	**Helicene Derivatives for Potential Optoelectronic and Spintronic Applications**
10:00 – 10:45	Abraham Nitzan	**Chirality of Molecular Vibrations and Its Dynamical Implications**
10:45 – 11:30	*COFFEE BREAK*	
11:30 – 12:15	**Poster Prizes**	
12:15 – 13:00	Concluding Remarks	
13:00	*LUNCH*	

End of the seminar and departure

For participants leaving on Friday a self-service breakfast will be provided on Friday morning.

Posters

Poster Session, Tuesday, 03 December, 16:30 h (CET)

Seif Alwan	**Spin-Polarized Transport and Chiral Spin Textures in the Chiral Inorganic Crystal CrNb${3}$S${6}$: Insights into the CISS Effect**
Jean-Philippe Ansermet	**Dissipative Spin-orbit Coupling**
Theo Balland	**Effects of Chiral Polypeptides on Skyrmion Stability and Dynamics**
Neeraj Bangruwa	**CISS-Controlled DNA Biosensor**
Mampi Biswas	**Thiol-Functionalized Chiral Molecules: Structure-Property Relationships, Enantioselective Surface Interactions, and CISS Effect**
Lilach Brann	**Non-classical Temperature Dependence of Chirality-Induced Magnetization and Its Implications for RNA’s Homochirality**
Arianna Cantarella	**Photoinduced CISS in Organic Molecules: Ab Initio Parametrization of a Generalized Hubbard Model**
Leonardo Celada	**Many-Body Models for Chirality-Induced Spin Selectivity in Electron Transfer**
Zinaida Charyshnikova	**Magnetic Field Effects on Spin Polarization in Tight-binding Models**
Namgee Cho	**Three-dimensional Electron Model for Chirality-induced Spin Selectivity**
Nicholas Davis	**Current-driven Mechanical Motion of Double Stranded DNA Results in Structural Instabilities and Chiral-induced-spin-selectivity of Electron Transport**

Poster Session, Tuesday, 03 December, 16:30 h (CET)

Tejas Deshpande	**Chiral Light-matter Interactions in Solution-processable Semiconductors**
Sumit Ghosh	**Chirality Induced Spin Selectivity from Charge Current**
Niccolò Giaconi	**Helicenes and Organic Radicals Monolayers on Gold: CISS Effect for Chiral Spintronics**
Alexander Hagenow	**Synthesis of Chiral Metallacrown Complexes and their Application as a Versatile Toolbox for the Investigation of Ciss-related Effects in Spintronics**
Olav Hellwig	**Microscopic and Macroscopic CISS Effect Characterization: Moving from MBE– to Sputter-Deposited Co/Au/Co Substrates**
Dominik Hornig	**CD Spectroscopy of Chiral Bismuth Oxido Nanoclusters and Their Use in Molecular Assemblies for Studies on the CISS Effect**
Aras Kartouzian	**Asymmetric Laser Desorption by Circularly Polarized Light**
Aadi Konidena	**Hubbard Interactions and the CISS Effect in Helical Molecules**
Sarthak Kumar	**Exploring Chiral-Induced Spin Selectivity (CISS) Effect: Unveiling Spin-Dependent Phenomena in Chiral Systems**
Thibaut Lacroix	**Comparison of Non-equilibrium Green Function and Wave Packet Methods for Simulating Spin-dependent Electron Transport**
Xuan Liu	**Chiral-related Interface Effects in Heterojunction Devices**

Poster Session, Tuesday, 03 December, 16:30 h (CET)

Offek Marelly	**Coherent Fluctuations in a Chiral Medium**
Marc Mende	**Synthesis of Chiral π-Conjugated Polymers for Optoelectronic Devices**
Olga Mironova	**Vanadyl-based Paddlewheels as Promising Molecular Qudits**
Ashish Moharana	**Vectorial Spin to Charge Conversion in Nanographenes**
Paul Valerian Möllers	**Probing the Roles of the Substrate, Temperature and Cooperative Effects in CISS**
Oliver Monti	**Surface Magnetic Stabilization and the Photoemission Chiral-induced Spin-selectivity Effect**
Vladimiro Mujica	**A Theoretical Model of the Connection Between Spin Polarization and Optical Activity in Chiral Molecules**
Sumit Naskar	**First-principles Approaches to Model Metal-chiral Molecule Interfaces to Understand the CISS Effect**
Thi Ngoc Ha Nguyen	**Influence of Molecular Electric Dipole Moment on Spin Polarization**
Berith Pape	**Putting a New Spin on Chiral Symmetry Breaking of Sodium Chlorate**
Jonah Peter	**The Chiral-Induced Spin Selectivity Effect of Light**
Ulrich Pototschnig	**First-principles Study of Chiral Spin Dynamics and Photoelectron Circular Dichroism**

Poster Session, Tuesday, 03 December, 16:30 h (CET)

Lokesh Rasabathina	**Microscopic and Macroscopic CISS Effect Characterization: Moving from MBE– to Sputter-Deposited Co/Au/Co Substrates**
Massimiliano Remigio	**Chirality-Induced Spin Selectivity at Solid-Liquid Interfaces: A Route to Enantioenriched Surface Growth**
Eva Rentschler	**Chiral Metallacrown Complexes**
Samuel Rudge	**The Role of Quantum Vibronic Effects in the Spin Selectivity of Charge Transport through Chiral Molecular Junctions**
Gaurav Sahu	**Chirality-Induced Spin Selectivity at the Single-Molecule Scale**
Franziska Schölzel	**Establishing Circular Dichroism Spectroscopy as Predictor for CISS in Molecular Thin Films**
Anil Kumar Singh	**Dual-Functionality in Single-Molecule Junctions: Exploring Electron-Chirality Interactions for Chiral Spintronics**
Julian Skolaut	**Influence of the Substrate on the Spatial Variation of Conductive Properties of Self-Assembled Monolayers**
Noah Smith	**Characterization of Chiro-Optical Properties and the Chirality-Induced Spin Selective Effect in Liquid Crystalline Semiconducting Systems**
Rudolf Smorka	**Influence of Nonequilibrium Vibrational Dynamics on Spin Selectivity in Chiral Molecular Junctions**
Simon Sochiera	**Spin Hall Magnetoresistance as a Probe for Chiral-Induced Spin Selectivity**

Poster Session, Tuesday, 03 December, 16:30 h (CET)

Dominik Steinmetz	**Efficient Implementation of the Berry Curvature in TURBOMOLE**
Johannes Strauch	**Towards First-Principles Electron—Phonon Coupling in Chiral Induced Spin Selectivity**
Adrian Urban	**Chirality-Dependent Electron Tunneling in the Small Molecule Limit**
Martin van Horn	**An Electronic Spin on Nuclear Dynamics Near Conical Intersections**
Ofek Vardi	**On the Origin of The Isotope Effects in Life**
Roland Winkler	**Categories of Chirality: Full Classification of Enantiomorphism**
Nir Yuran	**Chiral Molecular Coating of LiNiCoMnO 2 Cathode for High-Rate Capability Lithium-ion Batteries**

Abstracts of Lectures

(in alphabetical order)

Magnetic Field Effects and Spin-Correlated Radical Pairs in Chiral Donor–Bridge–Acceptor Molecules

Laura A. Völker[1] and John M. Abendroth[1]

[1]*ETH Zürich, Laboratory for Solid State Physics, Zürich, Switzerland*

Photogenerated spin-correlated radical pairs (SCRPs) are entangled electron spin pairs formed in well-defined spin states. Spin chemistry of SCRPs plays important roles in biology, having been famously implicated in the leading hypothesis for avian magnetoreception. At the same time, synthetic molecules that mimic these biologically relevant species such as donor–bridge–acceptor (D–B–A) compounds, which support SCRP formation via transfer of an electron from a donor species to a photoexcited acceptor, have gained attention for quantum information technologies. Recently, SCRPs in chiral systems, *e.g.*, D–B–A compounds with a chiral bridge, have received significant interest as ideal systems to study mechanistic aspects of CISS. Here, we show combined experimental and theoretical efforts toward probing the influence of bridge chirality on magnetic field effects and spin dynamics of SCRPs. First, we show characterization of magnetic field-dependent fluorescence attributed to charge recombination in chiral D–B–A molecules patterned as oriented monolayers. We then demonstrate using explicit spin dynamics simulations how all-optical assessment of SCRP triplet character as a hallmark of CISS may be experimentally realized. Lastly, we describe advancing efforts toward quantum sensing of CISS in SCRPs with single shallow nitrogen–vacancy (NV) centers in diamond. These include theoretical assessment of possible measurement schemes, experimental evaluation of NV photophysics under short wavelength excitation needed for photogeneration of SCRPs, and first detection of SCRPs in few molecule systems.

Enantioselective adsorption and spin-selective electron transport of discrete helicenes on cobalt

Daniel E. Bürgler

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Forschungszentrum Jülich, 52425 Jülich, Germany

Enantiospecific adsorption on magnetic surfaces and spin-selective electron transport through chiral molecules are major manifestations of chirality-induced spin selectivity (CISS). Despite the abundant experimental evidence gathered through mesoscopic adsorption and transport measurements, the exact mechanism behind CISS remains elusive. Here we report studies on CISS effects at the single-molecule scale performed under the well-defined conditions of surface science techniques, *i.e.* ultra-high vacuum, low temperatures, clean crystalline surfaces, and solvent-free deposition by sublimation [1]. It is shown that single helicene molecules undergo enantioselective adsorption on ferromagnetic cobalt surfaces. Spin- and chirality-sensitive scanning tunneling microscopy (STM) reveals that molecules of opposite handedness prefer adsorption onto cobalt islands with opposite out-of-plane magnetization. As mobility ceases in the final chemisorbed state, it is concluded that enantioselection must occur in a physisorbed transient precursor state. State-of-the-art spin-resolved ab-initio simulations support this scenario by refuting enantio-dependent chemisorption energies. These findings demonstrate that van der Waals interaction should also include spin-fluctuations, which are crucial for molecular magnetochiral processes [2]. Moreover, spin-polarized scanning tunneling spectroscopy enables the measurement of *I-V* curves through individual chemisorbed helicene molecules. Direct comparison of two enantiomers under otherwise identical conditions reveals magnetochiral conductance asymmetry or CISS-based magnetoresistance (CISS-MR) of up to 50% when either the molecular handedness is exchanged or the magnetization direction of the STM tip or Co substrate is reversed. The bias dependence of these asymmetries will be discussed. Overall, our single-molecule experiments rule out ensemble effects as well as electron–phonon coupling as primary mechanisms responsible for CISS in helicene molecules.

References

[1] M. R. Safari *et al.*, Nanomaterials **12,** 3281 (2022)
[2] M. R. Safari *et al.*, Adv. Mater. **36**, 2308666 (2024)
[3] M. R. Safari *et al.*, Small **20**, 2308233 (2024)

HELICENE DERIVATIVES FOR POTENTIAL OPTOELECTRONIC AND SPINTRONIC APPLICATIONS.

Jeanne Crassous

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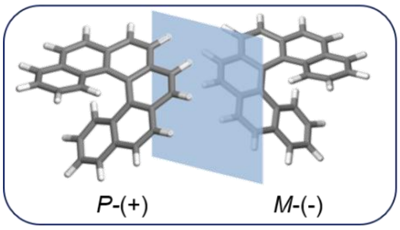
Ortho-fused aromatic rings form helically shaped chiral molecules such as carbo[6]helicenes, that wind in a left-handed (*M*) or a right-handed (*P*) sense.[1] The helical topology combined with extended π-conjugation provides helicenes with peculiar properties such as strong photophysical and chiroptical properties (high optical rotation values, intense electronic circular dichroism and circularly polarized emission). The molecular engineering of helicenes using organometallic and heteroaromatic chemistries offers a convenient way to tune the properties of these helically shaped □-ligands. Indeed, their combination with metallic or organic assembling units leads to chiral materials with appealing properties (circularly polarized phosphorescence, magnetochirality, spin selectivity) for applications in materials science (Circularly Polarized OLEDs, Chiroptical Switches, Spintronics). I will present a set of representative examples.[2]



- ***Strong circular dichroism***
- ***Circularly polarized emission***
- ***Chiral-induced spin selectivity***
- ***Magnetochirality***

References

[1] J. Crassous, I. G. Stará, I. Starý (Eds) *Helicenes - Synthesis, Properties and Applications.* Wiley (2022).

[2] a) E. S. Gauthier, L. Abella, N. Hellou, B. Darquié, E. Caytan, T. Roisnel, N. Vanthuyne, L. Favereau, M. Srebro-Hooper, J. A. G. Williams, J. Autschbach, J. Crassous, *Angew. Chem. Int. Ed.* **59**, 8394 (2020); b) M. Atzori, K. Dhbaibi, H. Douib, M. Grasser, V. Dorcet, I. Breslavetz, K. Paillot, O. Cador, G. L. J. A. Rikken, B. Le Guennic, J. Crassous, F. Pointillart, C. Train, *J. Am. Chem. Soc.* **143**, 2671 (2021); c) R. Rodríguez, C. Naranjo, A. Kumar, P. Matozzo, T.-K. Das, Q. Zhu, N. Vanthuyne, R. Gómez, R. Naaman, L. Sánchez, J. Crassous, *J. Am. Chem. Soc.* **144**, 7709 (2022); d) K. Dhbaibi, M. Grasser, H. Douib, V. Dorcet, O. Cador, N. Vanthuyne, F. Riobé, O. Maury, S. Guy, Amina Bensalah-Ledoux, B. Baguenard, Geert L. J. A. Rikken, C. Train, B. Le Guennic, M. Atzori, F. Pointillart, J. Crassous, *Angew. Chem. Int. Ed.* **62**, e202215558 (2023).

Surface magnetic stabilization and the photo-emission chiral-induced spin-selectivity effect.

Oliver L. A. Monti[1] and Yonatan Dubi[2]

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Department of Chemistry, Ben Gurion University of the Negev, 1 Ben-Gurion Ave, Beer Sheva, 8410501, Israel

The spinterface mechanism was suggested as a possible origin for the chirality induced spin-selectivity (CISS) effect [1], and was used to explain and reproduce, with remarkable accuracy, experimental data from transport experiments showing the CISS effect [2-5]. Here, we apply the spinterface mechanism to explain the appearance of magnetization at the interface between non-magnetic metals and chiral molecules. We show that the stabilization of surface magnetic moments occurs at a wide range of realistic parameters and is robust against dephasing. Importantly, we show that the direction of the surface magnetic moments is determined by the chiral axis of the chiral molecules. Then, armed with the concept of stable surface magnetic moments, we formulated a theory for the photo-emission CISS effect. The theory, based on spin-dependent scattering, leads to some direct predictions regarding the relation between the photo-emission CISS effect, the chiral axis direction, and the tilt angle of the detector with respect to the surface. These predictions are within reach for current experimental capabilities, and may shed new light on the origin of the CISS effect.

References

[1] S. Alwan and Y. Dubi, Spinterface theory for the chirality-induced spin-selectivity effect, J. Am. Chem. Soc. **143**, 35, 14235–14241 (2021)

[2] Y. Dubi, Spinterface chirality-induced spin selectivity effect in bio-molecules, Chemical Science **13**, 10878-10883 (2022).

[3] Yang, et al., Real-time monitoring of reaction stereochemistry through single-molecule observations of chirality-induced spin selectivity, Nat. Chem. **15**, 972–979 (2023).

[4] S. Alwan et al., Temperature-dependence of the CISS effect from measurements in Chiral molecular intercalation super-lattices, J. Chem. Phys. **159**, 014106 (2023).

[5] S. Alwan, A. Sharoni & Y. Dubi, Role of Electrode Polarization in the Electron Transport Chirality-Induced Spin-Selectivity Effect, J. Phys. Chem. C. 128 (15), 6438-6445 (2024).

A molecular perspective on the CISS effect

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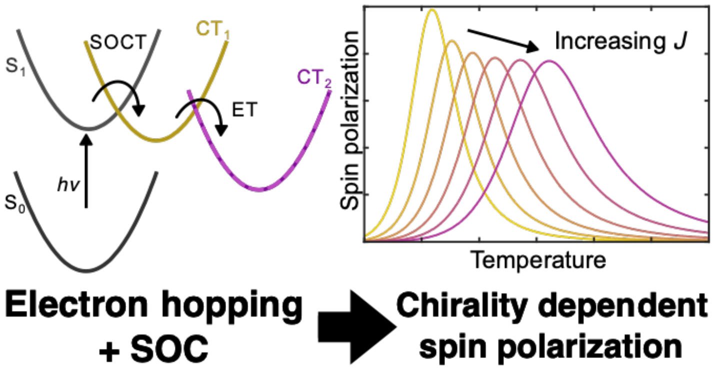
Much of the pioneering experimental and theoretical work the chirality induced spin selectivity effect has focussed on spin-dependent conductivity of chiral molecules between electrodes. Although instrumental in the development of our current understanding of the CISS effect, these set-ups are complicated by the interplay of effects arising from the chiral molecule directly and interactions with electrodes. Arguably a simpler set-up, which I will outline in this talk, is to consider photo-excitation induced electron transfer in chiral molecules, where the affect of chirality on electron transport can potentially be probed more directly.

In particular I will describe a theory of how electron spin coherence[1] and spin polarization[2] can be generated in chiral molecular systems in the absence of electrode interfaces. These effects emerge in systems where charge transport is dominated by incoherent hopping, mediated by spin–orbit and electronic exchange couplings. In this talk I will present the theory of these effects, and the general principles we can use to understand chirality induced spin effects in molecular systems. Furthermore, I will outline how chirality induced spin effects could be probed experimentally, and how it can also manifest in charge recombination reactions [3]. Chirality induced spin coherence and polarization effects should appear in many chiral systems, and the ideas I will present have implications for the study of spin transport in many systems.



References

[1] Fay, T. P. Chirality-Induced Spin Coherence in Electron Transfer Reactions. *J. Phys. Chem. Lett.* **12**, 1407–1412 (2021).
[2] Fay, T. P. & Limmer, D. T. Origin of Chirality Induced Spin Selectivity in Photoinduced Electron Transfer. *Nano Lett.* **21**, 6696–6702 (2021).
[3] Fay, T. P. & Limmer, D. T. Spin selective charge recombination in chiral donor–bridge–acceptor triads. *J. Chem. Phys.* **158** 194101 (2023)

Wave-function engineering on superconducting substrates:

Chiral Yu-Shiba-Rusinov molecules

Lisa Rütten[1], Harald Schmid[2], Eva Liebhaber[1], Giada Francesci[1], Ali Yazdani[1], Gaël Reecht[1], Kai Rossnagel[3], Felix von Oppen[2], Katharina J. Franke[1]

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[3] Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, 24118 Kiel, Germany

Exchange-coupled magnetic adatoms on superconductors induce Yu-Shiba-Rusinov (YSR) states within the superconducting energy gap of the substrate. The spatial extent and symmetry of the YSR states is influenced by the Fermi surface of the substrate and the local adsorption site, which imposes a crystal field of the adatom. Since the YSR states are protected by the superconducting energy gap, they provide an ideal platform for engineering states with intricate wave-function symmetries.

Here, we deposit Fe atoms on the quasi-two-dimensional superconductor 2H-NbSe$_2$ to engineer hybridization of YSR states. We use the tip of a scanning tunneling microscope (STM) to position the individual atoms in designated sites. We construct structures in which we deliberately break mirror symmetries, realizing symmetries that cannot be found in the orbitals of isostructural planar molecules in the gas phase. We exploit this potential by designing chiral YSR wave functions of triangular adatom structures.

Functional coordination compounds and the CISS effect

R. Rodrigues de Miranda,[1] I. Coloma,[2] M. Cortijo,[2] E. Hillard,[1] P. Rosa,[1] M. Gonidec[1]

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[2] Universidad Complutense de Madrid, Madrid, Spain

Functional molecular systems could find applications in a broad range of technological systems. To take advantage of their, over the years, many efforts have focused on grafting a variety of organic molecules and some interesting coordination compounds comprising ad-hoc modifications to allow to bind them specifically to a substrate of choice for the envisioned applications. Most often, this is done by using sulfur based anchoring groups on coinage metals or alternate anchoring groups (phosphonic acids, carboxylic acids…) on metal oxides. Our group focuses on the design and study of such functional coordination compounds for a wide range of applications going from molecular electronics and spintronics to quantum computing. Indeed, coordination compounds are highly versatile systems that present multiple options for tuning their electronic structure and thus their physical properties making them targets of choice for physical-organic studies on surfaces.

In this presentation, we will first outline the work that we have performed in recent years to successfully graft a variety of switchable Fe (II) Spin crossover (SCO) complexes that retain their properties on metallic surfaces and that open interesting avenues for switchable CISS systems. We will also outline recent efforts to study the CISS effect in large-area molecular junctions of chiral Ru paddlewheel coordination complexes on surfaces.

Fine-Tuning ORR Activity at Chiral Imprinted Mesoporous Metal Surfaces

Zikkawas Pasom[1,2], Jonas Fransson[3], Chularat Wattanakit[1]* and Alexander Kuhn[1,2]*

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[2] University Bordeaux, CNRS, Bordeaux INP, ISM UMR 5255, 33607 Pessac, France
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The Chiral-Induced Spin Selectivity (CISS) effect has been proposed as an interesting concept for many different applications [1,2]. In particular, it significantly impacts the kinetics of the oxygen reduction reaction (ORR), which plays an essential role in clean energy production, such as fuel cells, due to the higher electron transfer efficiency from chiral electrode surfaces to oxygen molecules [3]. To date, this effect has been mostly investigated for electrodes modified with chiral organic monolayers or chiral assemblies. However, the CISS effect at intrinsically chiral metal surfaces has not been studied so far. Recently, we have successfully developed chiral imprinted Pt-Ir alloys with mesoporous features by electroreduction of metal salts in the simultaneous presence of non-ionic surfactants and various chiral compounds, such as the enantiomers of phenyl ethanol [4,5]. These designer materials can perfectly retain molecular chiral information, even after template removal. This allows them to be used for various applications, such as asymmetric synthesis of chiral compounds and enantioselective analysis.

In this contribution, chiral imprinted mesoporous Pt-Ir has been studied as an electrocatalyst for ORR. We show that such electrodes can significantly improve ORR when imprinted with one type of enantiomer. However, this effect cannot be observed when using an achiral electrode of analog composition, or the one imprinted with the opposite enantiomer, which, in strong contrast, seems to inhibit ORR. This work opens up novel perspectives for developing catalytic systems for ORR, promoted by intrinsically chiral metal surfaces.

References

[1] Mtangi, W.; Kiran, V.; Fontanesi, C., Naaman, R., J. Phys. Chem. Lett., **6**, 4916 (2015).

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Analyzing CISS Mechanisms through Spin-Dependent Dispersion Relations

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Chirality-induced spin selectivity (CISS) is an electron spin-filtering effect induced by chiral molecules, such as DNA and helical proteins. Although this effect has been demonstrated in various experimental setups, the underlying mechanism of CISS remains unclear. In this talk, I will present our studies on several CISS models, where we investigate the mechanisms using a combination of wave packet method and analysis based on spin-dependent dispersion relations.

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How to use Chiral or Magnetic Fields to Boost Clean Energy Processes

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Achieving a sustainable energy future requires a critical shift from fossil fuels to renewable energy sources. Central to this transition is the advancement of efficient catalysts capable of converting chemical energy into electricity or utilizing electrons to drive chemical transformations. Recent *in-situ* and *operando* studies at electrified interfaces have achieved remarkable progress, offering molecular-level insights and control over catalytic processes at the atomic scale [1]. However, the influence of electron spin on the activity and selectivity of catalytic interfaces remains largely underexplored.

In this talk, I will show how applying magnetic fields [2] or utilizing chiral self-assembled monolayers [3–5] can substantially enhance the efficiency and selectivity of 2D catalysts. By leveraging spin-polarized currents, we can optimize the performance of Earth-abundant catalysts, offering a pathway to reduce our dependence on critical materials for energy applications. This approach highlights the potential of spin-selective phenomena in maximizing catalytic effectiveness and contributes to the broader goal of finding sustainable, critical-element-free energy solutions.

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Microscopic origin of magnetoresistance in chiral molecule/ferromagnet junctions

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Chirality is defined as a pseudo-scalar that changes sign under mirror reflection and is characterized by the lack of mirror symmetry. Recently, numerous reports have highlighted various phenomena associated with chirality-induced spin selectivity (CISS) [1,2]. Notably, these CISS-related phenomena include the manifestation of magnetoresistance in junctions that involve chiral molecules and ferromagnetic electrodes. However, theoretical frameworks suggest magnetoresistance should not occur within the linear response regime [3]. Despite this, experimental studies have reported exceptionally large magnetoresistance, exceeding those observed in commercially available spintronic devices, such as the CoFeB/MgO system. Consequently, achieving a comprehensive and unified understandings of CISS-related phenomena remains a significant challenge, hindering practical applications and further advancements in this field. In this study, we used the prototypical chiral electrolyte, (1S)-(+)- or (1R)-(−)-camphor-10-sulfonic acid (S- or R-CSA) [4], in combination with ferromagnetic electrodes, to perform time-resolved observation of magnetoresistance and discuss its microscopic origins [5]. We will explore the role of spin polarization in chiral molecules, which seems related to recent observation of CISS-related phenomena occurring without a bias current in the system, suggesting thermally driven spin polarization in chiral molecules [6-8].

This is a work in collaboration with T. Yamamoto and T. Nozaki of AIST, T. Nagata, S. Sakamoto, M. Shiga, W. Gao, K. Inoue of the University of Tokyo, and T. Takenobu of Nagoya University. This work was partially supported by JSPS-KAKENHI, JST-ASPIRE, Spin-RNJ, and MEXT-Xnics (JPJ011438).

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CISS- more surprises to come

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The chiral induced spin selectivity (CISS) effect was discovered more than two decades ago, but its studies keep producing interesting and sometimes unexpected results. Among them the establishing the role of chiral phonons in the effect [1] and the ability to transfer spin information through chiral materials using those phonons [2]. Other findings, discovered very recently, have to do with the ability to pump heat using CISS and the coherent properties of electrons passing through chiral systems. Some of these effects will be presented and discussed in relation to the mechanism of the CISS effect.

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Chirality of molecular vibrations and its dynamical implications

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The suggestion that chiral phonons are implicated in the CISS phenomenon is attractive given several possibly related magnetic and optical phenomena involving nuclear motions in chiral systems as well as accounts of the temperature dependence of such phenomena. This talk will present our published (below) and some unpublished studies aimed at understanding the nature of chiral molecular vibrations, quantifying their chirality, and studying their manifestations in molecular transport phenomena.

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First-principles study on the electron-vibration and spin-phonon couplings of chiral molecules

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Chirality induced spin selectivity (CISS) [1,2] is a unique phenomenon which possesses potential applications for efficient energy-conversion devices, while its mechanism is not fully revealed. Presenter has been studying electrical and thermoelectrical properties of molecular junctions with theoretical calculations and has an interest in spin-related phenomena [3-5]. It is known that first-principles calculations are not able to quantitatively predict the chirality-induced magnetoresistance. Here, the effect of molecular vibrations of (1S)-(+)- or (1R)-(−)-camphor-10-sulfonic acid (S- or R-CSA) on its spin density was investigated by first-principles calculations including spin-orbit coupling and non-colinear spin [6]. Infrared vibrational circular dichroism (VCD) and electron-vibration coupling were extracted for each vibration mode. Along with some vibration modes having both strong electron-vibration and chiral-phonon couplings, change in spin densities were calculated. The results suggest thermally driven spin polarization in chiral molecules[7-9].

This is a work in collaboration with T. Yamamoto and T. Nozaki of AIST, T. Nagata, S. Sakamoto, M. Shiga, W. Gao, K. Inoue of the University of Tokyo, K. Kimura of Osaka metropolitan Univesity, Y. Yamamoto of Institute of Molecular Science, and T. Takenobu of Nagoya University. This work was partially supported by JSPS-KAKENHI (22K18320, 22H00290, 22H04964, and 24H02234), JST-ASPIRE (JPMJAP2317), JST-PRESTO (JPMJPR2115) Spintronics Research Network of Japan (Spin-RNJ), and MEXT-Xnics (JPJ011438).

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Chiral spintronics using chiral - chiral interactions

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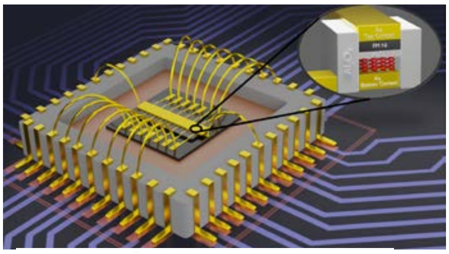

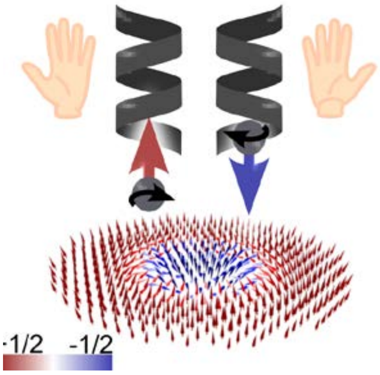

Chiral spintronics technology has the potential to overcome the limitations of other magnetic-based memory technologies and to facilitate the fabrication of inexpensive, high-density memory and other spintronics elements [1]. Recently, by utilizing the CISS effect we demonstrated a simple magnetless spin based magnetic memory [2-5]. When chiral molecules are adsorbed on the surface of thin ferromagnetic film, they induce magnetization perpendicular to the surface (the MIPAC effect) [5].

Utilizing the MIPAC and chiral-chiral interactions enables to reduce memory size stabilizing chiral spin textures in magnetic thin films by tuning skyrmion properties. Chiral molecules can locally manipulate magnetic properties by inducing magnetization through spin exchange interactions and by creating spin currents. The same effects were demonstrate using non-collinear antiferromagnetic thin films of Mn3Sn and chiral gating to achieve local, efficient antiferromagnetic memory.

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Ab initio spin dynamics and decoherence in solids

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Ab initio spin dynamics and transport simulations are critical for predicting new materials and realizing the potential of spintronics, spin-based quantum information science, and spin-selective photo-chemistry. In particular, these would be invaluable to predict key physical parameters including spin lifetime, spin diffusion and coherence length, magneto-optical spectra, and (spin)-photocurrent.

In this presentation we will introduce our recently developed real-time density-matrix dynamics approach with first-principles electron-electron, electron-phonon, electron-impurity scatterings and self-consistent spin-orbit coupling[1]. We are developing a computational framework and an open-source implementation for simulating spatio-temporal quantum dynamics and transport accounting for a range of quantum degrees of freedom (e.g., charge, spin, orbital, lattice).

We show our methods can accurately predict spin and carrier lifetime, spin diffusion length, and pump-probe Kerr-rotation signatures for general solids, with examples of Si, GaAs, 2D materials, and hybrid halide perovskites[2]. In particular, we show our recent study of how *g* factor fluctuations (including orbital angular momentum) lead to spin dephasing in halide perovskites under external magnetic field, and we will show the distinct electron-phonon contributions to spin and carrier relaxations and crucial dependence on crystal symmetry[2].

We next will introduce our recent progress of developing methodology for spin-optotronic signatures, such as circular dichroism and circular/spin photogalvanic effect to chiral and broken-inversion-symmetry solid[3].

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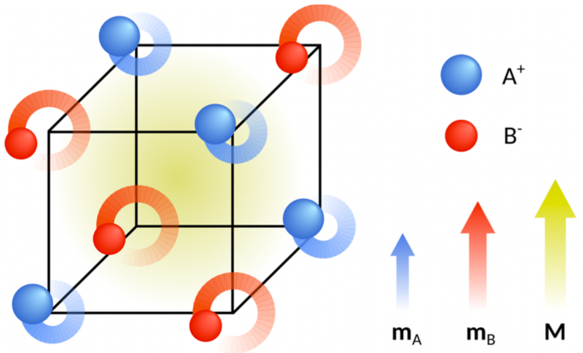
Have we learned anything from studying chiral phonons that might be relevant for CISS?

Nicola Spaldin

ETH Zürich

The emerging field of *chiral phonons* is an area of rapidly expanding research activity, with both theoretical predictions and experimental observations of novel behaviors associated largely with the angular momentum of the phonons. Of particular interest are the phonon magnetic moments, which should be at the limit of detectability based on simple models but have been found to yield unexpectedly large Zeeman splittings and magnetooptical Kerr responses suggesting that they are much larger than anticipated. This happy situation of the experimental magnetic response resulting from the chirality being much larger than the theoretically predicted behavior is perhaps reminiscent of the situation in current-induced spin selectivity. In this talk I will provide a review of recent progress in the field of chiral phonons (in which I have been active) and try to draw some analogies with the field of chiral induced spin selectivity (in which I am a beginner and will need your help) in the hope of making some potentially useful connections.



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Ab Initio Modelling of Chirality Induced Spin Selectivity

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Chirality induced spin selectivity (CISS), i.e. the ability of a material or single molecule to selectively transport electrons of a given spin orientation, has been identified as a promising approach to initialise and read out molecular spins for quantum information processing.[1] However, studying the molecular origin of the CISS effect is still in its early stages. Recent breakthroughs have revealed the potential for direct detection of the CISS effect in electron donor-acceptor dyads where a chiral thiabridged[4]helicene (TAHEL) acts as the donor and perylenediimide (PDI) as the acceptor (see Figure 1). They are covalently linked through bridges of different lengths.[2]

Ab initio modelling presents a powerful tool to enhance the understanding, e.g. by assessing the phonon-assisted nature of the CISS mechanism in the absence of strong spin-orbit coupling. Moreover, accurate calculations are suited to drive molecular design towards highly selective spin filters. In this work, promising CISS candidate molecules are modelled from first principles by employing density functional theory (DFT) methods for the description of ground state properties such as molecular geometry and vibrations, while correlated wavefunction methods are used to model the electronic structure of the low-lying spin states including spin-phonon coupling as well as spin-orbit effects into the calculation. To simulate molecular properties under realistic and experimentally testable conditions, molecular systems are further studied in condensed phase environments, e.g. in periodic matrix embeddings or adsorbed to the surface of a periodic substrate. Preliminary calculations on the thia[4]helicene-perylenediimide dyads will be presented.

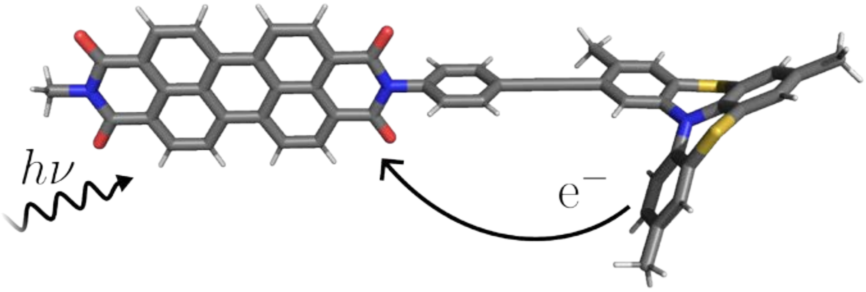

Figure 1: Photoinduced electron transfer in a thia[4]helicene-perylenediimide dyad.

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The mechanism of the molecular CISS effect in chiral nanojunctions

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The transmission properties of electrons through chiral systems attracts currently a lot of attention. In DNA and polypeptides the so-called chiral induced spin selectivity (CISS) effect describes the built-up of an extraordinarily high spin-polarizations upon long-range electron transfer. Recently, we studied PA molecules adsorbed on surfaces of magnetic Al2O3/Pt/Au/Co/Au nanostructures with perpendicular anisotropy. Thereby, a correlation between the PA molecules ordering at the surface with the electron tunneling across this hybrid system as a function of the substrate magnetization orientation as well as the coverage density and helicity was observed. The highest spin polarization values were found for ordered self-assembled monolayers and with a defined chemical coupling of the molecules to the magnetic substrate surface, showing that the current induced spin selectivity is a cooperative effect [1,2]. In a recent experiment the same molecules were adsorbed only the Au-tip [3]. Surprisingly, the magnetizations of the Co layer had to be oriented in opposite directions, e.g, for a high electron transmission. The result clearly shows that the orientation of the intrinsic electric dipole, which for PA increases with the length of the molecules, plays a decisive role. The approximately point-symmetric behavior of the IV curves can be understood by a spin polarized interface, induced and defined by the helicity and electric dipole orientation of the molecule at the interface. Even more, only a low spin-orbit coupling in the molecule allows high transmission of spin-polarized and non-polarized electrons and explains similar IV curves for the two configurations. In this sense, the helical system does not act as a simple spin filter or polarizer.

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Influence of electronic-vibrational coupling on spin-selectivity in chiral molecular junctions

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Chirality-induced spin selectivity (CISS) describes the phenomenon that the transport of electrons through a chiral molecule depends on the spin polarization of the electrons. The CISS effect has been measured in a variety of setups, however, a detailed understanding of the underlying mechanisms is still elusive [1]. Theoretical studies have pursued different mechanisms, including many-body effects induced by electron-electron interaction or electronic-vibrational coupling [2]. In this contribution, we investigate the role of the coupling of the electrons to the molecular vibrations in transport scenarios where the molecule is coupled to leads [3]. The study uses the hierarchical quantum master equation approach HEOM [4,5] to describe transport processes and considers different models of electronic-vibrational coupling in chiral molecules [6,7]. The results show that the influence of electronic-vibrational coupling depends on the specific model and the physical parameters. The underlying mechanisms are analyzed in some detail.

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Mechanism for Electrostatically Generated Magnetoresistance in Chiral Systems without Spin-Dependent Transport

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Electrical studies of chirality induced spin selectivity usually involve a chiral system which is brought into contact with a ferromagnetic electrode or ferromagnetic tip. A magnetoresistance (MR) is observed, which depends on the magnetization direction (relative to the surface) and the sign of the chirality. In recent years these experiments have also been performed in the linear transport regime, where a change in the slope of the I/V characteristics is observed, when either the magnetization direction or the chirality is changed.

Some years ago we indicated that these observations are puzzling, because they are in contradiction with the reciprocity theorem for two-terminal measurements, which imply that there should be no change in the electrical resistance when the magnetization direction is reversed [1,2]. A detailed analysis of the spin and chirality dependent transport coefficients shows that in a chiral system spin dependent transmission has to be accompanied with spin flip reflection. A consistent analysis of spin and charge transport confirms the absence of MR in chiral -ferromagnet systems. I will show that in the linear regime an MR can occur in a chiral system due to Hanle spin precession due to an applied magnetic field, however with inverted sign as compared to the conventional Hanle effect with ferromagnetic electrode [3].

We have also shown that CISS type magnetoresistance can occur in the non-linear electron transport regime [2]. However the sign of the MR was found to reverse when the voltage bias is reversed. This disagrees with the usual experimental observation that the CISS MR does not change sign with bias. As a possible solution we have now proposed an electrostatic mechanism for the generation of a MR in chiral systems, which does not require spin dependent transport [4]. We conjecture that the electrostatic potential at the interface between the chiral system and ferromagnet depends on the interplay of chirality and magnetization direction. In our mechanism the MR therefore arises from a change in the charge transport, and not the spin transport. However, as I shall discuss, we currently have no insight in the possible microscopic origins of this electrostatic effect.

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Electronic Transport Across Chiral Molecules

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Over the past decade, there has been tremendous progress in the measurement, modeling and understanding of structure-function relationships in single molecule circuits. Experimental techniques for reliable and reproducible single molecule junction measurements have led, in part, to this progress. In particular, the scanning tunneling microscope-based break-junction technique has enabled rapid, sequential measurement of large numbers of nanoscale junctions allowing a statistical analysis to readily distinguish reproducible characteristics. Although the break-junction technique is mostly used to measure electronic properties of single-molecule circuits, in this talk, I will demonstrate its use to measure conductance and current-voltage characteristics of chiral molecules using a magnetized scanning tunneling microscope substrate and a standard gold tip. I will present results of our measurements over a series of molecules that include both conjugated and saturated systems.

Experimental Investigations of Spin and Charge Transport in Chiral Materials

David H. Waldeck

Department of Chemistry, University of Pittsburgh, Pittsburgh PA 15260 USA

We describe recent experimental investigations of spin-polarized charge currents and pure spin currents in chiral materials, including chiral metal oxides, chiral quantum dots, and chiral polymer fibers. We will discuss the implications of this work for the mechanism of electron spin selectivity in chiral matter.

Chiral-induced unidirectional spin-to-charge conversion

A. Wittmann

Johannes Gutenberg University Mainz, Germany

The observation of spin-dependent transmission of electrons through chiral molecules has led to the discovery of chiral-induced spin selectivity (CISS). The remarkably high efficiency of the spin polarizing effect has recently gained substantial interest due to the high potential for future sustainable hybrid chiral molecule magnetic applications. However, the fundamental mechanisms underlying the chiral-induced phenomena remain to be understood fully. In our recent work, we explore the impact of chirality on spin angular momentum in hybrid metal/ chiral molecule thin film heterostructures [1]. For this, we inject a pure spin current via spin pumping and investigate the spin-to-charge conversion at the hybrid chiral interface. Notably, we observe a chiral-induced unidirectionality in the conversion. Furthermore, angle-dependent measurements reveal that the spin selectivity is maximum when the spin angular momentum is aligned with the molecular chiral axis. Our findings validate the central role of spin angular momentum for the CISS effect, paving the path toward three-dimensional functionalization of hybrid molecule-metal devices via chirality.

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The spin of photoelectrons from monolayers of helical molecules

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We report about results for the longitudinal spin polarization of photoelectrons released from (sub-) monolayers of small helical molecules adsorbed on metal surfaces. Deep-UV laser radiation at λ = 213 nm was employed, which yielded kinetic energies of the photoelectrons up to about 1.5 eV. The electron spin polarization was detected by Mott scattering, thereby integrating over the spectrum of the kinetic energy. The molecular systems studied were [7]-helicene, dithia-[7]-helicene, and Ni-tetrapyrrole. The molecules yielded a spin polarization of the photoelectrons even for excitation with linearly polarized light and with values up to about P = 35 %. Such high values allowed to investigate the dependence of the spin polarization on various external parameters, like surface temperature and surface density. For all adsorbed molecules no variation of the spin polarization was observed when varying the temperature in a wide range. Changing the density of the molecules yielded another result. For tetrapyrrole the spin polarization increased in a way which can be associated with the conclusion that in this molecule the CISS effect is a single molecule effect. Thus cooperative effects do not play a role. For dithiahelicene a similar result was found for a large range of surface coverage, up to about 0.85 ML. For a full monolayer, however, a significant increase of the value of the spin polarization was observed. That can either be explained by a re-arrangement and a phase transition at this high density or with signs for a cooperative effect.

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Abstracts of Posters

(in alphabetical order)

Dissipative spin-orbit coupling

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The thermodynamics of irreversible processes [1] was applied for a system composed of electric and magnetic dipoles. The Onsager method, well-known to account for cross-effects such as the coupling between heat and electricity, is applied to this case, and leads to a dissipative coupling [2] between the evolution of the electric and magnetic dipoles. To understand this result, the electric field is calculated for a charge going down a helix. While the resulting spin-orbit coupling resulting from this type of calculation is very weak, it can be argued that fluctuations associated with molecular vibrations may provide a relaxation mechanism for the spins, along the helix axis. This relaxation corresponds to the dissipative coupling predicted by thermodynamics.

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Effects of chiral polypeptides on skyrmion stability and dynamics

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CISS, chirality-induced spin selectivity is a phenomenon that has raised significant interest due to the large spin polarizations generated by organic light element molecules and other effects such as magnetic switching of ferromagnets induced by chiral molecules[1]. Chiral polypeptides like alpha-helix polyalanine (AHPA) are an example of molecules that exhibit the CISS effect. In hybrid systems, these chiral molecules have been observed to influence magnetic properties such as changes in the magnetization[1]. In our work, chiral molecules are found to increase the pinning of magnetic domains[2]. Our objective is to investigate how these chiral molecules interact with chiral spin structures, namely magnetic skyrmions, which are stabilized in ferromagnetic/heavy metal multilayers due to Dzyaloshinskii-Moriya interaction[3]. In particular, we explore the effect of the chiral molecules absorbed on skyrmions-hosting multilayers. Using magneto-optic Kerr effect imaging, we show that chiral polypeptides can influence the stability of skyrmions by modifying the spin reorientation transition temperature and the range of the applied magnetic field in which they are stable. We also show that the chiral molecules affect the skyrmion dynamics, in particular the thermal diffusion of the skyrmions.

Acknowledgments

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CISS-Controlled DNA Biosensor

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The Chiral-induced Spin Selectivity (CISS) effect is a remarkable phenomenon with the capability to distinguish between the spin of electrons as they pass through chiral molecules by backscattering one of the spin components. In this work, we explore the role of the CISS effect in time-controlled single photon counting (TCSPC) to detect DNA hybridization. We observe that the average lifetime of optical excited states of quantum dots attached to double-strand (ds) DNA varies with the directions of the magnetic field. Specifically, the difference in the nonradiative average decay lifetime is 2206 ps (~2.2\ ns) in the case of hybridized strands, which is 130 times higher than that observed with quantum dots attached to single-stranded DNA. On the contrary, this difference becomes almost negligible in the case that hybridization fails. Additionally, we explore the use of Fourier Transform infrared spectroscopy for the detection of double-strand DNA in the presence of the magnetic field. We provide a theoretical framework to support the experimental evidence of magnetic field-dependent FTIR spectroscopy for the detection of dsDNA.

Keywords: CISS effect, Time-controlled single photon counting, DNA hybridization, FTIR.

Thiol-Functionalized Chiral Molecules: Structure-Property Relationships, Enantioselective Surface Interactions, and CISS Effect

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The chiral induced spin selectivity (CISS) [1] effect links the handedness of chiral molecules to their spin-polarization properties. When these molecules interact with spin-polarized, ferromagnetic surfaces, they exhibit spin-dependent enantioselective adsorption. One enantiomer preferentially interacts due to the alignment of its intrinsic spin with the surface's spin polarization. The molecular dipole moment plays a crucial role in determining the polarization's sign, emphasizing the dipole's orientation relative to the surface. Our study explores how the structure-property relationships of chiral compounds influence the CISS effect. Examining molecular dipole moments, chemical environments, and stereoisomeric configurations deepens our understanding of chirality's role in spin-selective surface interactions, aiding in designing chiral molecules for selective interaction with spin-polarized surfaces through chemisorption and dimerization. In this work, we have synthesized thiol-bearing chiral molecules with good yields and high enantiomeric excess (*ee* > 97%) by varying dipole moments for studying their interaction with spin-polarized Au-Ni surfaces using techniques such as chiral high-performance liquid chromatography (HPLC) with chiral stationary phase, scanning tunneling microscopy (STM) and electrochemical quartz-crystal microbalance (EQCM).[2] By characterizing the adsorption kinetics and behavior of these chiral molecules, we aim to provide insights into the enantioselective properties and potential applications of CISS. Furthermore, electron-rich, or electron-deficient chiral molecular systems exhibiting the pillow effect[3] on metal surfaces can form self-assembled monolayers (SAMs) on Au-Ni surfaces. These SAMs can be utilized as source and drain electrodes for organic field-effect transistors (OFETs). [4] Such chiral molecular systems are highly valuable for enantiomeric separation in pharmaceutical and industrial applications.

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Photoinduced CISS in organic molecules: ab initio parametrization of a generalized Hubbard model

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In this work, we present an *ab initio* study of a chiral molecule previously shown to exhibit the Chirality-Induced Spin Selectivity (CISS) effect upon Photoinduced Electron Transfer.[1] Using DFT and TD-DFT, we investigate the electronic ground-state and excitations, finding good agreement with experimental absorption spectra. Our results reveal a weak electronic coupling between the donor, chiral bridge and acceptor subunits of the chiral molecule, allowing for an independent study of each subunit. Considering recent developments suggesting a significant role of electron correlations in CISS [2,3], we model the chiral bridge using a multi-orbital generalized Hubbard model. To parameterize the Hubbard model, we employ an *ab initio* strategy, extracting parameters through CASCI calculations. This framework provides a versatile approach that may be extended to other systems. Additionally, as supported by previous research [5,6], we investigate low-energy vibrations as a potentially crucial factor for reproducing the CISS effect in electron transfer. Through torsional potential energy scans across key dihedral angles, we observe notable asymmetry in the chiral bridge and assess the impact of these torsions on the inter-site Hubbard parameters.

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Many-Body Models for Chirality-Induced Spin Selectivity in Electron Transfer

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The phenomenon of chirality-induced spin selectivity (CISS) has drawn significant attention for its potential applications in molecular electronics and spin-based quantum devices, but its underlying mechanisms remain inadequately understood, especially regarding the precise influence of molecular structure. We propose a microscopic model that explicates CISS in electron transfer (ET) through short chiral chains by accounting for internal many-body interactions within the chiral bridge [1]. This model allows us to derive solutions that clarify the emergence of spin polarization on the acceptor site and the factors influencing this effect. Many experiments suggest that CISS is particularly sensitive to vibrational interactions within the molecular bridge [2], a factor critical in our model, where both spin-orbit coupling (SOC) and vibrational modes interactively influence coherent and incoherent ET dynamics, creating sustained spin polarization. Our study highlights the fundamental role of electron-electron correlations, which enable the formation of distinct many-body states on the bridge and substantially enhance SOC effects, yielding significant spin polarization levels. Employing a Schrieffer-Wolff transformation, we derive an effective Hamiltonian in which vibrational coupling boosts spin selectivity, aligning with recent observations of CISS in donor-acceptor molecular systems [3]. These findings advance the theoretical framework around CISS mechanisms, suggesting new perspectives for investigating chirality-based spin phenomena. This project is supported by the Horizon Europe program through the ERC-Synergy CASTLE project (proj. n.101071533).

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Magnetic field effects on spin polarization in tight-binding models

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Quantum dynamics offers crucial insights into particle and spin behavior that cannot be provided by classical mechanics.[1] Following breakthroughs in understanding the quantum nature of matter, the electronic and spin characteristics of nanoscale objects became accessible to quantitative study, including the modeling of circular dichroism and optical rotatory dispersion, the spin Hall effect, and the Rashba effect. Our research focuses on the chiral induced spin selectivity (CISS) effect, which remains poorly understood. The CISS effect describes spin polarization emerging from electron transport through chiral systems.

Several theoretical frameworks have been proposed to explain spin polarization in the CISS effect, including scattering models[2], substrate interaction models[3], and models based on topological spin phase effects[4]. The topological spin phase effect suggests that the absence of inversion symmetry in chiral systems creates an effective magnetic field that influences electron spins, even in the absence of an applied magnetic field[5]. This framework provides the starting point of our investigation, where we develop tight-binding models to explore magnetic field effects on electron spin polarization.

Our findings indicate that a magnetic field influences spin polarization only at extremely high field values, greater than ~ 10^3 Tesla. Ongoing studies are exploring how spin polarization changes with even and odd numbers of orbitals in the model and how the oscillation time of spin polarization varies as we change the structure’s orientation with respect to the field. These investigations aim to explore how magnetic fields may contribute to the CISS effect.

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Three-dimensional electron model for chirality-induced spin selectivity

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Chirality-induced spin selectivity (CISS) is an electron spin filtering effect in which electrons moving through chiral molecules become spin-polarized. To unravel the mechanism behind CISS, various theoretical models have been investigated, with spin-orbit couplings induced by the electric fields of chiral molecules considered a key factor. However, in most theoretical studies, electron motion has been approximately described using low-dimensional tight-binding models where electrons are assumed to propagate along pre-determined one-dimensional helical paths, and the electric fields acting on the electrons have a specific helical symmetry of choice. In reality, both electron propagation and spin-orbit couplings are determined by the electrostatic potentials of the chiral molecules. In this work, we explore whether and how CISS effects emerge in a three-dimensional electron model, where electrons move through a three-dimensional electrostatic potential with chiral symmetry, and investigate how spin polarization depends on the handedness and length of the chiral potential.

Current-driven mechanical motion of double stranded DNA results in structural instabilities and chiral-induced-spin-selectivity of electron transport

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Chiral-induced-spin-selectivity of electron transport and its interplay with DNA’s mechanical motion are explored in a double stranded DNA helix with spin–orbit-coupling. The mechanical degree of freedom is treated as a stochastic classical variable experiencing fluctuations and dissipation induced by the environment as well as force exerted by nonequilibrium, current-carrying electrons. Electronic degrees of freedom are described quantum mechanically using nonequilibrium Green’s functions. Nonequilibrium Green’s functions are computed along the trajectory for the classical variable taking into account dynamical, velocity dependent corrections. This mixed quantum-classical approach enables calculations of time-dependent spin-resolved currents. We showed that the electronic force may significantly modify the classical potential, which, at sufficient voltage, creates a bistable potential with a considerable effect on electronic transport. The DNA’s mechanical motion has a profound effect on spin transport; it results in chiral-induced spin selectivity, increasing spin polarization of the current by 9% and also resulting in temperature-dependent current voltage characteristics. We demonstrate that the current noise measurement provides an accessible experimental means to monitor the emergence of mechanical instability in DNA motion. The spin resolved current noise also provides important dynamical information about the interplay between vibrational and spin degrees of freedom in DNA.

Chiral light-matter interactions in solution-processable semiconductors

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Chirality is a property which is widely observed in nature and refers to the characteristic that an object cannot be superimposed on its mirror image. Recently, chiral solution-processable semiconductors have seen a surge of interest for their potential applications in emerging photonic, optoelectronic and spintronic technologies [1]. Yet, the underlying chiral light-matter interactions, especially in the excited state, which lead to spin and light polarization remain poorly understood [2].

Here, we present our preliminary results on using transient chiroptical spectroscopy to understand the consequences of morphological (hierarchical) chirality for charge, spin and light polarization. We compare achiral chromophores in solution and a solid-state matrix with their arrangement into chiral twisted molecular crystal films with large chiral domains. Our results reveal complex photophysics and unexpectedly even point towards persistent excited-state chirality induced by circularly-polarized excitation.

By studying transient chiral light-matter interactions we hope to aid in understanding the mechanistic origin of chiral-induced spin-selectivity (CISS) and leverage such spin polarization for efficient emission of circularly polarized light from cheap solution-processable semiconductors.

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Chirality induced spin selectivity from charge current

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We present a theoretical model of chirality induced spin selectivity in chiral molecule based on the charge dynamics. The proposed mechanism does not require the presence of any intrinsic spin-orbit coupling and therefore is well suited for molecules made of light atoms like C. We systematically show that how the structural chirality controls the flow of charge in these molecules which eventually leads to their spin selectivity. Finally, we show how this model can explain the salient features of recent experiment on spin-selective electron transport through single chiral molecules [1].

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Helicenes and Organic Radicals Monolayers on Gold: CISS effect for Chiral Spintronics

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Organic materials have traditionally been used as charge transport agents in spintronics, while inorganic materials have dominated data processing and storage. However, the discovery of the Chirality Induced Spin Selectivity (CISS) effect[1] has widened this field, revealing that chiral organic molecules can exhibit effective spin selectivity at room temperature. In this context, we previously reported the deposition of a monolayer of enantiopure helicene radical cations on gold via non-covalent interactions.[2] Looking for a more stable system, we now present a novel thioacetyl derivative of thia-bridged triarylamine helicene, along with its corresponding radical cation, designed for direct chemisorption on a Au(111) surface. After monolayer deposition, we performed thorough surface characterization, supported by theoretical calculations, to gain insight into the molecular arrangement on the gold surface. To further investigate the CISS effect, we fabricated a micrometric device embedding a monolayer of these molecules. The device revealed an enantiomer-dependent asymmetric trend in magnetoresistance (MR), confirming the CISS effect. Additionally, magnetic conductive - Atomic Force Microscopy (mc-AFM) showed efficient electron spin filtering at room temperature, even at low potentials.[3] Importantly, the radical functionality on the molecular backbone did not suppress the spin polarization of the transmitted current.[4] These findings underscore the potential of thiahelicenes as key candidates for the development of advanced novel CISS-based technologies.

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SYNTHESIS OF CHIRAL METALLACROWN COMPLEXES AND THEIR APPLICATION AS A VERSATILE TOOLBOX FOR THE INVESTIGATION OF CISS-RELATED EFFECTS IN SPINTRONICS

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Metallacrowns (MCs) are a group of polynuclear, macrocyclic coordination compounds. Their name is derived from crown ethers, organic macrocycles, which are able to coordinate a metal ion inside their central cavity [1]. MCs share this feature with their organic counterparts, but in addition to the capability of incorporating transition-metal ions as well as lanthanides, the surrounding macrocycle itself contains multiple transition metal ions. This leads to a multitude of interesting magnetic phenomena, ranging from tunable magnetic exchange interactions between ring metals and the central ion [2] to single molecule magnetic behavior [3]. Another feature of MCs is their planarity and stability. The combination of those features with their magnetic properties has made MCs the focus of intense research in the field of spintronics. With the rising interest in the impact of the CISS effect on spintronic applications, e.g. skyrmions and their behavior [4], we are investigating different strategies to introduce chirality into the MCs. Thereby we want to make the vast number of MCs available for research related to the CISS effect.

Herein, we present three different strategies that have been successfully pursued to introduce chirality into MCs. In all cases, the different effects of the chirality on the structural and magnetic properties of the MCs have been investigated. It can also be shown, that MCs are a suitable platform to develop chiral, single molecule magnets.

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Microscopic and Macroscopic CISS Effect Characterization: Moving from MBE– to Sputter-Deposited Co/Au/Co Substrates

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High spin polarization in alpha-helical polyalanine molecules enables selective electron transport with a defined spin direction, a phenomenon known as Chirality Induced Spin Selectivity (CISS) [1]. This discovery holds promising implications for organic spintronic devices. Furthermore, the adsorption of pure enantiomers of α-helical polyalanine on a gold-covered ferromagnetic thin film, termed Magnetism Induced by the Proximity of Adsorbed Chiral molecules (MIPAC), can influence the magnetization of the ferromagnetic thin film. In our ongoing research, we are combining microscopic STM/STS and macroscopic MOKE magnetometry of hybrid spin-valve structures for CISS effect characterization of Au/Co/Au tri-layer systems. Microscopically we observe magneto resistance changes across the hybrid spin-valve structure as we reverse the Co layer with an external magnet[2]. Furthermore, we measure the HOMO/LUMO gap via STS measurements. Macroscopically we find a coercivity enhancement of the Co layer reversal after exposure to homochiral molecule solution[3]. To use more complex magnetic substrate layer systems in the future within hybrid spin-valve structures, we moved from MBE– to sputter deposited substrate systems and discuss the challenges that are connected with this transition.

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CD spectroscopy of chiral bismuth oxido nanoclusters and their use in molecular assemblies for studies on the CISS effect

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Circular Dichroism (CD) Spectroscopy is a well-established technique for distinguishing enantiomers in chiral materials. CD spectroscopy, which measures the differential absorption of left- and right-handed polarized light, can predict the strength of the chirality-induced spin selectivity (CISS) effect.[1,2] Currently, we follow a concept, which makes use of atomically precise and thus monodisperse metal oxido nanoclusters (MO-NCs) of bismuth (BiO-NCs) as potential materials to induce hyperpolarization upon excitation. The BiO-NCs show one of the largest diversities with various sizes of the [Bi_xO_y] cluster core in combination with an exchangeable ligand shell. The latter has been demonstrated for the quite stable [$Bi_{38}O_{45}$] cluster architecture and enables targeted modification of the BiO-NC properties such as the introduction of chirality. By functionalizing of such BiO-NCs with amino acids like Boc-L-methionine, chiral nanoclusters [$Bi_{38}O_{45}$(Boc-L-Met-O)$_{24}$] can be synthesized and studied in solution using CD spectroscopy.[3–5] This technique can also be applied to investigate ligand exchange reactions of different amino acids with BiO-NCs, which forms the basis for the direct binding of the BiO-NCs to helical chiral peptides for further investigation of the CISS effect. Further the tuning of physical properties will be demonstrated by doping these atomically precise nanoclusters with other metals.

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Asymmetric laser desorption by circularly polarized light

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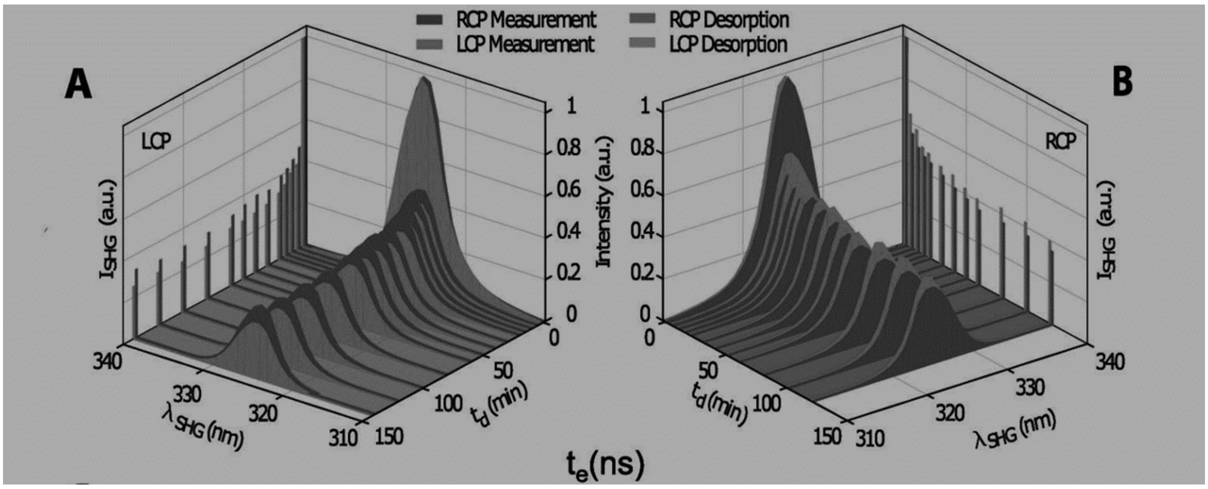
The interest in enantioseparation and enantiopurification of chiral molecules has been drastically increasing over the past decades, as these are critical steps in fields such as the pharmaceutical industry, asymmetric catalysis, and chiral sensing. By exposing racemic samples of BINOL (1,1 '-bi-2-naphthol) coated onto achiral glass substrates to circularly polarized (CP) light, we have observed that the molecules desorb with different efficiency from the surface depending on the handedness of the CP light (*1, 2*). This phenomenon demonstrates that preferential desorption of enantiomers can be achieved by controlling the handedness of CP light, though the current understanding of laser desorption does not quantitatively explain this effect. One possible explanation involves the CISS (Chirality-Induced Spin Selectivity) effect, suggesting that the interaction of CP light with the achiral substrate may lead to the ejection of spin-polarized electrons, which interact preferentially with chiral adsorbates, thus inducing a preferred excitation-induced desorption. Our findings, together with a simplified phenomenological model, indicate that the process of laser desorption needs further development, and the role of quantum mechanical processes should be revisited to account for these data. Asymmetric laser desorption offers a contamination-free technique for enantioenrichment of chiral compounds (3).



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Hubbard Interactions and the CISS Effect in Helical Molecules

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Chirality Induced Spin Selectivity is an exciting phenomenon where the geometry of helicoid molecules evidently determines the resulting electron spin when a current is driven through the molecule[1]. Despite experimental backing, the details of the primary mechanism that drives spin selection is still not understood. In fact, theoretical simulations based on spin-orbit coupling alone still disagree substantially with experimental data, prompting the investigation into alternate rationales for the observed spin-polarised current. To assess the possible role of the electron-electron correlation effects in current spin polarisation, we study the quantum transport through a molecular junction with Hubbard interactions added on each site of a helical molecule alongside the spin-orbit coupling. We employ the Non-Equilibrium Green's Function (NEGF) formalism and Feynman diagram technique to account for the Hubbard interactions within the Second Born approximation. To calculate the spin current as a function of the temperature and the applied bias, our treatment goes beyond the simple Landauer approach and incorporates the Hubbard self-energies in the equation for the full current.

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Exploring Chiral-Induced Spin Selectivity

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Chiral Induced Spin Selectivity (CISS) is a phenomenon in which chiral molecules or materials induce spin-polarized currents when subjected to an electric field. This effect has gained significant attention in the field of spintronics, where the manipulation of electron spin is used to enhance the performance of electronic devices. In this study, we explore the CISS effect in various chiral organic materials, focusing on their potential applications in spintronic devices such as spin filters, sensors, and memory storage elements. Using both experimental measurements and theoretical models, we demonstrate that the spin polarization is highly dependent on the handedness of the chiral molecules, with right- and left-handed structures exhibiting distinct spin polarization characteristics.

Our results also show that the CISS effect can be tuned by varying the molecular environment, temperature, and applied external fields. We discuss the underlying mechanisms of CISS, including the role of spin-orbit coupling and electronic structure in chiral molecules. These findings open new avenues for designing spintronic devices that utilize organic materials with high spin polarization and low power consumption, providing insights into the future of flexible and efficient spintronic technologies.

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Comparison of non-equilibrium Green function and wave packet methods for simulating spin-dependent electron transport

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Non-equilibrium Green function (NEGF) technique has been widely employed to simulate quantum transport at the molecular scale [1]. It is one of the main methods used to study transport properties of chiral molecules which are central to the investigation of chiral induced spin selectivity (CISS). Equivalent to the solution of a scattering problem, this approach leads to an energy resolved description of transport properties that can be used to obtained spin-dependent currents.

Alternatively, the description of electronic transport can be performed at the level of the Schrödinger equation. In that case, one considers an initial wave packet in the position basis – which therefore corresponds to a superposition of energy eigenstates – and computes its dynamics under the total Hamiltonian of the molecule and the leads. One benefit of this approach is to readily give access to the transient dynamics of the electron inside of the molecule, thus opening the possibility to study the internal mechanisms underlying CISS.

We demonstrate that NEGF results can be recovered from wave packet evolution and that tailored wave packets enable to access directly specific transport properties of interest [2]. These results show that while they rely on different formalisms wave packet evolution and NEGF do give the same information about the response of the system.

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Chiral-related interface effects in heterojunction devices.

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The interface between materials usually plays an important role on the device properties, and chiral factors make the properties of heterojunction interfaces containing chiral materials more diversified.

Chiral heterojunctions formed by two adjacent chiral molecules is common in electronic devices, and experiments show that this structure can be used for enantiomeric separations. By considering the chiral-induced spin-orbit coupling (SOC) determined by the geometric structure of the molecule, we found that the spin-related interface couplings of the homochiral and heterochiral configurations are different. Further calculating their transport properties using the Landauer-Büttiker equation, we found that the current of the homochiral configuration is greater than that of the heterochiral configuration. Based on this, the concept of chiral resistance is proposed and the influence factors such as the molecule length and the chirality mismatch are discussed. [1]

We also construct a heterojunction composed by a chiral molecule and achiral metal layers, and propose a mechanism of chirality transfer from chiral molecules to metals by the interface coupling. It is found that the transferred charges present a chiral distribution even in the achiral metal. Considering the strong SOC of the metal, the induced chirality in the metal provides an additional chiral channel with strong SOC for the moving electrons, which is conducive to inducing a significant spin polarization. [2]

In conclusion, the spinterface and chiral interface in devices containing chiral materials have significant impacts on electron transmission, which could be utilized in the development of future electronic and spintronic devices.

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Coherent Fluctuations in a Chiral Medium

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The CISS effect refers to the ability of chiral materials to act as spin filters for electron transport [1]. While much work has been done in terms of studying the CISS effect and its exact origin, these have focused mostly on direct currents, whereas alternating currents (AC) have remained mostly unexplored. This work explores what happens when the currents driven through chiral mediums are alternating, particularly when the period of the driven currents is shorter than the spin lifetime. If the period is shorter than the spin lifetime, the spin polarized current should not have time to lose its polarity between current cycles. This should lead to a rectifying effect with spins of different polarities accumulating at the different edges of the chiral medium. In this work, to study the impact of AC currents, we shall drive high frequency currents through chiral mediums of various widths and measure the voltage response as a function of frequency. Furthermore, by utilizing the spin Hall effect, we shall measure the spin lifetime in the different chiral mediums. Once the period of the driven current is equal to the spin lifetime, the electrons will not have time to lose their spin polarity, and we expect to see the appearance of Hall voltage. These measurements will be followed by high frequency experiments to determine whether the spin lifetimes can be manipulated and what is the performance and behavior of the CISS effect at high frequencies. These results will shed new light on the origin and different components that make up the CISS effect, from an unexplored direction. Aside from the contributions to our understanding of the CISS effect, the results of this work will greatly contribute to the development of reliable CISS-based spintronic devices with high operating frequencies. Limited spin lifetime is a major obstacle in the development of reliable spintronic devices. Thus, preventing or delaying the loss of spin polarity is critical for the advancement of this field [2].

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Synthesis of Chiral *π*-Conjugated Polymers for Optoelectronic Devices

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This project focuses on the synthesis and characterization of novel chiral π-conjugated polymers for application in circularly polarized organic light-emitting diodes (CP-OLEDs) and spintronic devices employing the CISS effect. Building upon previous work, that identified a "sweet spot" in the ratio of chiral components for optimal circular dichroism (CD) and electroluminescence dissymmetry factors (g_{EL}), the research employs a dual-strategy approach, combining the "Sergeant and Soldier" principle with intrinsic chirality in polymer design. Key objectives include the synthesis and optimization of chiral π-conjugated polymers by fine tuning the molecular structure as well as the ratio of chiral components incorporated into the polymer chain. Based on former high performing polymer structures, this approach should result in high CD and g_{EL} values with overall favourable optoelectronic properties for CP-OLEDs and CISS devices. Current progress includes implementing and refining synthetic procedures, designing new chiral π-conjugated polymers and conducting preliminary analysis of chiroptical properties. Using these chiral polymers as a 1D template for other high-performance achiral conductive polymers achieving chiral supramolecular organization could also lead to more efficient and versatile organic optoelectronic devices. The projects findings will contribute to the development of improved CP-OLEDs and provide valuable insights for the broader field of organic electronics and spintronics.

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Vanadyl-based paddlewheels as promising molecular qudits

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Over the past two decades, the CISS effect has manifested mainly in electrochemistry, water splitting, and photoluminescence. Recently, it has been recognized as a possible key resource in quantum information technologies based on electron spin, since it allows achieving spin polarization at RT and potentially eliminates the need for sub-kelvin temperatures. At the same time, molecular spin qubits and qudits can be integrated into (supra)molecular CISS-active architectures and exploited as coherent and highly sensitive magnetic sensors for a better understanding of the CISS effect. Both proposals are at the core of ERC SYNERGY project CASTLe (no. 101071533).

The molecular route to creating spin qubits and qudits is less used than leading solid-state approaches, but not less promising. An accurate design of coordination compounds enables achieving T_2 values approaching milliseconds, satisfying the requirement for quantum gate operations. Among suitable compounds, vanadyl-based complexes stand out thanks to consistently long T_2 and easily interpreted response, having an $S = 1/2$ electron spin coupled to the nuclear spin of ^{51}V ($I = 7/2$), which yields an electronuclear spin qudit.

Recently, vanadyl-based paddlewheels (PWs) [$\mathbf{Pt}VO(SCOR)_4$] (R = Me, Ph) were investigated as a new family of qudits within the CASTLe project. The vanadium cation in these complexes is surrounded by nuclear spin-free oxygen atoms, and the monothiocarboxylate ligands comprise only H atoms as a source of decoherence from nuclear spins. PWs are potentially processable, e.g., by deposition exploiting metallophilic interactions with the PtS_4 moiety, and modifiable by changing the ligand environment. To open access to a broader chemical modification, we decided to explore similar but previously unknown Pd-based PWs, [$\mathbf{Pd}VO(SCOR)_4$] (R = Me, Ph). In addition, we created isostructural diamagnetic titanyl-based analogs, serving as host materials for solid-state magnetic dilution experiments.

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Probing the Roles of the Substrate, Temperature and Cooperative Effects in CISS

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Monolayers of heptahelicene ([7]H) and helical tetrapyrroles (TPBT) were adsorbed onto different single-crystalline metal substrates, and the spin polarization (SP) of photoelectrons emitted from these surfaces was measured as a function of the temperature and the surface coverage.

We demonstate that full monolayers of either molecule can generate a significant photoelectron SP exceeding $P = 30\%$. This finding is attributed to the CISS effect because the sign of the SP is reversed between both enantiomers of the molecules. The CISS-related SP values measured upon excitation with linearly polarized light are largely independent of the atomic number and the electronic structure of the substrate material.

The temperature-dependent population of those vibrational modes which affect the helical molecular geometry was calculated via DFT. Combined with these results, the temperature-dependent SP data suggest that molecular vibrations do not play a significant role for CISS in these molecules.

To investigate the possible role of intermolecular interactions in the emergence of CISS, we measured the SP at varying submonolayer coverages. The SP measured on the TPBT layers scales nonlinearly with the surface coverage. However, we demonstrate that this behavior can be rationalized entirely through changes of the photoelectron yield upon surface functionalization, and that it therefore represents no evidence for cooperative effects in CISS. Corresponding data measured on [7]H layers, in contrast, indicate a strong increase in the SP at a surface coverage between 84% and 100% of a monolayer. We hypothesize that this reflects a phase transition in the molecular ordering within the layer.

Chiral Induced Spin Selectivity Effect at Helical Nanographene-metal Interfaces

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The observation of spin-dependent transmission of electrons through chiral molecules has led to the discovery of chiral-induced spin selectivity (CISS). The high efficiency of the spin filtering effect in chiral molecules has recently gained significant interest due to the high potential for novel hybrid molecule magnetic spintronics applications. However, the fundamental mechanisms underlying the CISS effect at the molecule-metal interface remain incompletely understood. In our work, we explore spintronic phenomena at hybrid nanographene and metal interfaces to elucidate the underlying mechanisms of the CISS effect. This study focuses on the influence of chiral helical nanographene adsorption on spin-to-charge conversion at the molecule-metal hybrid interface. Our results show that the molecular orientation plays a crucial role in the chiral induced spin polarization effect of nanographene, paving the path toward the three-dimensional engineering of hybrid interfaces.

Surface magnetic stabilization and the photoemission chiral-induced spin-selectivity effect

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The spinterface mechanism was suggested as a possible origin for the chirality induced spin-selectivity (CISS) effect, and was used to explain and reproduce, with remarkable accuracy, experimental data from transport experiments showing the CISS effect. Here, we apply the spinterface mechanism to explain the appearance of magnetization at the interface between non-magnetic metals and chiral molecules, through the stabilization of otherwise fluctuating magnetic moments. We show that the stabilization of surface magnetic moments occurs for a wide range of realistic parameters and is robust against dephasing. Importantly, we show that the direction of the surface magnetic moments is determined by the chiral axis of the chiral molecules. Armed with the concept of stable surface magnetic moments, we then formulate a theory for the photoemission CISS effect. The theory, based on spin-dependent scattering, leads to direct predictions regarding the relation between the photoemission CISS effect, the chiral axis direction, the spinterface ”size”, and the tilt angle of the detector with respect to the surface. These predictions are within reach of current experimental capabilities, and may shed new light on the origin of the CISS effect.

A theoretical model of the connection between spin polarization and optical activity in chiral molecules

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Abstract: Chiral structures exhibit non-zero chiroptical activity (COA) due to the coupling between their electric and magnetic responses under external electromagnetic fields, an absent effect in achiral systems, where space-inversion symmetry is preserved. Non-magnetic chiral structures also exhibit Chiral-Induced Spin Selectivity (CISS), primarily detected in molecular junctions, where spin selection emerges without external magnetic influence. Despite the different origins of these physical phenomena, our model captures the relevant physics required to address the experimentally measured connection between them, through the analysis of a predictor, connecting the CISS and COA responses, that depends on the magnitude of the spin polarization.

In a recent article (1), we derived explicitly how the electronic system couples with polarized electromagnetic radiation to yield a spin-dependent polarization Rosenfeld tensor, predicting characteristic spin signatures in the COA. Our model also predicts that under some conditions the COA will be affected by the spin-orbit interaction in a way that resembles recent findings about surface-induced symmetry breaking in the COA.

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First-principles approaches to model metal-chiral molecule interfaces to understand CISS effect

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The chiral induced spin selectivity (CISS)[1] effect considers the trajectory of electronic motion coupled to the orbital motion connected through spin-orbit coupling (SOC). It has implications in various fields of spintronics, catalysis and biology[1,2]. To elucidate the effect of SOC, we have employed two-component density functional theory (DFT). To make CISS relevant to applications, it is also important to achieve sujbstantial conductance. Conductance for conjugated chiral macrocycles were modeled using density functional tight-binding (DFTB) method. Our results suggest that SOC has an essential effect in spin selection in the chiral structures[3,4] and from DFTB results we can predict higher conductance for macrocycles showing higher chiroptical activities.

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Influence of Electric Dipole Moment on Spin Polarization

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Recent studies have demonstrated the significant impact of electric dipole moment orientation in chiral molecules on spin polarization[1]. When chiral molecules with strong dipole moments adsorb onto a surface, their adsorption breaks symmetry at the interface, resulting in enhanced spin-polarized currents. Conversely, chiral molecules with weak dipole moments exhibit only minimal spin polarization. Our findings hightlight the crucial relationship between the strength and orientation of electric dipole moments in chiral molecules and their effect on spin polarization, particularly at the interface.

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Putting a New Spin on Chiral Symmetry Breaking of Sodium Chlorate

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Imagine a world where we could harness the intrinsic spin of the electron to selectivity create one specific enantiomer. Transforming a simple chemical process into a high-precision dance choreographed by the very spin of the electron. The pursuit of uncovering new approaches for obtaining pure enantiomers is of great importance in the pharmaceutical industry and other chemical entities. In this study, for the first time, we explore how the chiral induced spin selectivity (CISS) effect can enable chiral symmetry breaking of crystallizing sodium chlorate ($NaClO_3$) as a new deracemization technique. Deracemization is a highly sensitive process and can easily be manipulated[1]. It is therefore the perfect subject to study the CISS effect.

$NaClO_3$, although achiral in solution, forms enantiomorphous crystals as it crystallizes as a conglomerate in the chiral space group $P2_13$[2]. The *l*- and *d*-crystals can easily be identified by a pair of polarizers. When two polarizers are crossed, the crystal's color, depending on the handedness of the crystal, either turns lighter or darker when rotating the filter clockwise or anticlockwise. In this process, we initiate chiral symmetry breaking by the specific interaction between the aligned spins in ferromagnetic substrates and the spin in $NaClO_3$ leading to the selective crystallization favoring one enantiomorph over the other[3,4]. This initial small imbalance is amplified to produce predominantly single enantiomorphic crystals of $NaClO_3$.

Our preliminary results hint to an enantiospecific deracemization of $NaClO_3$ induced by the aligned spins of the ferromagnetic substrate. A strong correlation between the magnetic field direction and the handedness of the crystals was observed. Evidently, when the spins were aligned down an enantiomeric excess of *d*-crystals was detected, while with the spin up an enantiomeric excess of *l*-crystals was observed. These results represent an experimental demonstration of deracemization brought about by the aligned spin within the ferromagnetic substrates, and not solely by the magnetic field. Developments such as these may open new frontiers in chemistry.

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The Chiral-Induced Spin Selectivity Effect of Light

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The chiral-induced spin selectivity (CISS) effect has eluded a consensus theoretical explanation but continues to drive tremendous progress in spintronics, materials science, electrochemistry, and molecular biology. Here we show that there exists a similar phenomenon for light in which photons of opposite circular polarization experience the characteristic properties of CISS. We demonstrate that dissipation, though not essential for breaking the spin degeneracy, plays a critical role in generating macroscopic spin polarizations. Our work clarifies the role of spin-dependent dissipation in CISS, thereby elucidating the room-temperature nature of the effect. These results pave the way for innovative photonics platforms exploiting strong chiral light-matter interactions, with potential applications in quantum information processing, optical devices, and next-generation communication technologies.

Chiral phosphoric acid-based self-assembled monolayers for spintronic applications

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The chirality-induced spin selectivity (CISS) effect opens up the possibility of employing organic molecules in spintronic applications. Aside from previously published results for thiol-based self-assembled monolayers (SAMs), phosphonic and phosphoric acid SAMs may well become significant for those CISS applications that require molecular coupling to oxide surfaces. We have studied the spin selectivity of both enantiomers of an organophosphoric acid derivative. These low molecular mass, aromatic chiral compounds were grown on metal oxides as about 1 nm thick SAMs. X-ray photoelectron spectroscopy and atomic force microscopy (AFM) was used to confirm the formation of distinct monolayers. To assess the spin selective behavior of these molecules, they were deposited onto the native oxide of a 100 nm thick Nickel substrate. First measurements using magnetic-conductive AFM indicate a spin polarization in excess of 80%. Due to its small size, compatibility with oxide surfaces, and relative ease of synthesis, the molecule is appealing for the realization of novel (nanoscale) organic spintronic devices.

First-principles study of chiral spin dynamics and photoelectron circular dichroism

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Understanding electron dynamics in chiral structures on ultrashort timescales is an important step towards understanding chirality- dependent processes in chemistry and biology [1]. Most notably, chiral molecules can induce spin polarization in an electronic current passing through them, known as the chiral induced spin selectivity effect (CISS), the theoretical understanding of which remains elusive [2]. Recent advances in attosecond research have now opened the door to capture electron-driven chiral dynamics of molecules in real time using time-resolved photoelectron circular dichroism (TR-PECD) measurements [3]. TR-PECD uses a pump-probe setup, where a linearly polarized pump pulse excites an electron wave-packet, and a circularly polarized probe pulse induces ionization after a well-defined time delay. Resulting electronic currents demonstrate an enantiosensitive response with periodic modulations as a function of pump-probe delay [4]. We employ real-time time-dependent density functional theory to simulate TR-PECD spectra for (R)/(S)-methyl lactate. In addition, the photoelectron currents are examined for time- and chirality-dependent spin behaviour.

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Microscopic and Macroscopic CISS Effect Characterization: Moving from MBE– to Sputter-Deposited Co/Au/Co Substrates

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High spin polarization in alpha-helical polyalanine molecules enables selective electron transport with a defined spin direction, a phenomenon known as Chirality Induced Spin Selectivity (CISS) [1]. This discovery holds promising implications for organic spintronic devices. Furthermore, the adsorption of pure enantiomers of α-helical polyalanine on a gold-covered ferromagnetic thin film, termed Magnetism Induced by the Proximity of Adsorbed Chiral molecules (MIPAC), can influence the magnetization of the ferromagnetic thin film. In our ongoing research, we are combining microscopic STM/STS and macroscopic MOKE magnetometry of hybrid spin-valve structures for CISS effect characterization of Au/Co/Au tri-layer systems. Microscopically we observe magneto resistance changes across the hybrid spin-valve structure as we reverse the Co layer with an external magnet[2]. Furthermore, we measure the HOMO/LUMO gap via STS measurements. Macroscopically we find a coercivity enhancement of the Co layer reversal after exposure to homochiral molecule solution[3]. To use more complex magnetic substrate layer systems in the future within hybrid spin-valve structures, we moved from MBE– to sputter deposited substrate systems and discuss the challenges that are connected with this transition.

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Chirality-Induced Spin Selectivity at Solid-Liquid Interfaces: A Route to Enantioenriched Surface Growth

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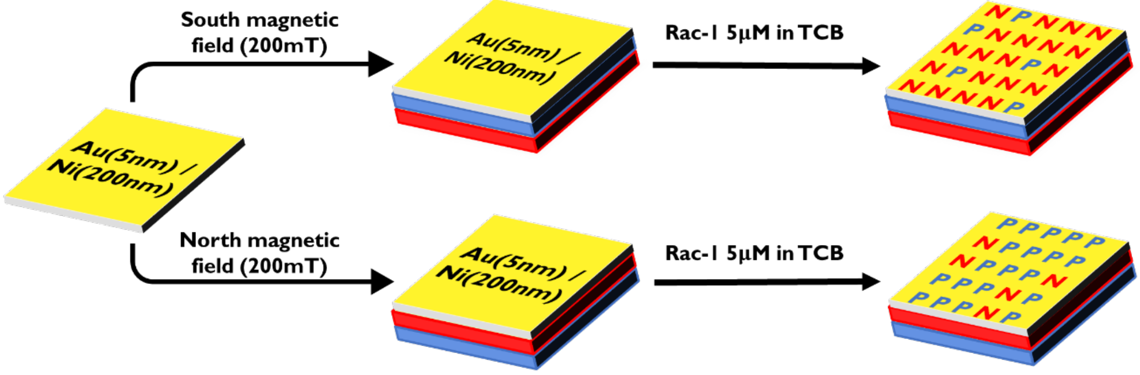
The chirality-induced spin selectivity (CISS) effect, which links electron spin to the handedness of chiral molecules, has emerged as a promising phenomenon with applications in quantum science, spintronics, and enantioselective processes.[1] Earlier studies using low-temperature scanning tunneling microscopy (LT-STM) for chiral resolution of racemic mixtures revealed limitations associated with vacuum conditions and low temperatures.[2] To address these challenges, we explored the self-assembly of chiral paracyclophane (PCP) molecules on ferromagnetic substrates at solid-liquid interfaces. The use of solvents at room temperature more closely mimics real-world conditions and promotes dynamics and growth of enantioenriched surfaces. Our experiments achieved an enantiomeric excess (ee) of over 70%, highlighting the potential of the CISS effect for generating enantioenriched surfaces and offering new possibilities for cost-effective and widely applicable enantioseparation technologies.



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Chiral Metallacrown Complexes at interfaces

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With the increasing interest in the implications of the CISS phenomena for spintronic applications, one of the crucial tasks is to investigate the impact of the adsorption of chiral magnetic molecules on the interfacial density of states and the corresponding magnetic properties of both the molecules and the substrate surface.[1-3] We have designed molecules with different geometries to introduce chirality into molecular magnetic materials. In particular, we focus on the unique chemical and structural features of molecules in the metallacrown family. Metallacrown complexes (MCs), which consist of metal ions and bridging ligands that self-assemble into cyclic ring structures, offer a particularly promising approach. Their atomic configuration allows tunable magnetic exchange between the metal ions within the ring structure, providing the basis for improving the properties of SMMs by incorporating ions with pronounced magnetic anisotropy. In an innovative approach, we combine the synthesis of magnetically interesting metallacrowns with advanced methods to study the molecular systems on the surface.

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The Role of Quantum Vibronic Effects in the Spin Selectivity of Charge Transport through Chiral Molecular Junctions

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Although the exact mechanism underpinning the chirality-induced spin selectivity (CISS) effect is still unknown, one of the leading ideas is that it is connected to the interactions between the transport electrons and molecular vibrations. In this work, we follow this theme by investigating CISS in the context of charge transport through a chiral molecular nanojunction via the numerically exact, fully quantum hierarchical equations of motion (HEOM) approach. Given that HEOM is numerically exact and treats all degrees of freedom quantum mechanically [1], our approach extends previous investigations into the connection between CISS and molecular vibrations, which rely on an approximate treatment of the vibrational part. Specifically, we calculate charge currents through a model with two electronic sites and two vibrational modes, for which significant spin selectivity has already been reported in the adiabatic, high-voltage regime [2]. In contrast, the HEOM approach is able to access the highly nonadiabatic regime of off-resonant charge transport, in which we find significant spin selectivity.

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Chirality-Induced Spin Selectivity at the Single-Molecule Scale

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Chirality-induced spin selectivity (CISS) causes spin-selective electron transport in chiral molecules and enantiospecific adsorption on magnetic surfaces. Here, these CISS effects are reported for single chiral heptahelicene molecules ([7]H) sublimed in UHV onto perpendicularly magnetized Co nanoislands on Cu(111) [1]. Spin-polarized scanning tunneling microscopy and spectroscopy (SP-STM/STS) at low temperature (5 K) is used (i) to determine for each individual [7]H molecule its handedness and the magnetization direction of the underlying Co-nanoisland and (ii) to measure spin-polarized electron transport through individual and separated [7]H molecules. Statistical analysis of 740 of molecules on 107 islands provides unequivocal evidence for enantioselective adsorption, *i.e.* molecules of opposite handedness prefer adsorption onto Co islands with opposite out-of-plane magnetization. As mobility ceases in the final chemisorbed state, it is concluded that enantioselection must occur in a physisorbed transient precursor state [2]. Furthermore, *I-V* curves of two enantiomers under otherwise identical conditions show magnetochiral conductance asymmetry and CISS-based magnetoresistance (CISS-MR) of up to 50% at 5 K when either the molecular handedness is exchanged or the magnetization of the STM tip or Co-substrate is reversed [3]. Our results show that CISS is a single-molecule effect and rule out ensemble effects as well as electron-phonon coupling as primary mechanisms for CISS.

Additionally, our future initiatives aimed at advancing the study and observation of CISS through temperature-controlled measurements or using iso-electronically substituted tetrapyrroles, will be the points of discussion.

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Establishing Circular Dichroism Spectroscopy as predictor for CISS in molecular thin films

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Circular Dichroism Spectroscopy is a well-established method in the field of chiral materials especially in chemistry and biology used to distinguish between the two enantiomers. It has been shown that the differential absorption of left and right circularly polarized light is able to predict the strength of the chirality induced spin selectivity (CISS) effect [1]. A high CD signal can hint towards strong spin filtering properties [2] whereas the sign of the CD directly correlates with the sign of the spin polarization [3]. Although CD spectroscopy has proven itself as a powerful tool it is mainly used for the analysis of molecules in solution. However, in many studies CISS relies on the interface between a chiral molecule and a magnetic substrate [1]. Therefore, it is crucial to develop a method for the interpretation of CD exhibited by molecular thin films.

In this study we present a homebuilt setup for the Jasco J-1500 CD spectrophotometer to implement the analysis of thin films systems into the device. Furthermore, we propose a way to overcome artefacts that may be caused by Linear Dichroism and Linear Birefringence related to the anisotropy of the film and the substrate [4]. We demonstrate our findings on systems of different chiral molecules like polypeptides, amino acids and bismuth-oxido nanoclusters.

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Dual-Functionality in Single-Molecule Junctions: Exploring Electron-Chirality Interactions for Chiral Spintronics

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Electron interactions within chiral media are very relevant to fields such as drug separation, chemical synthesis, and electronic transport across chiral materials, from bulk conductors to single molecules. Despite their significance, understanding these interactions at the atomic scale remains challenging. This study investigates electron-chirality interactions in single-molecule junctions based on individual helicene molecules suspended between ferromagnetic and non-ferromagnetic metal electrodes. These molecular junctions display dual functionality, acting as both a magnetic diode and a spin valve. Detailed analysis indicates that the magnetic diode behavior is driven by Electrical Magneto-Chiral Anisotropy (EMCA), while the spin valve effect arises from Chirality Induced Spin Selectivity (CISS). This coexistence of distinct interactions within a single nano system not only shed new light on electron-chirality interactions at the atomic scale, but also offers new opportunities to enhance the variability of spintronics by combining these unique functionalities in one device.

Further, a comparative magneto transport study of single-molecule junctions, with and without ferromagnetic electrodes, indicates that ferromagnetic electrode plays a crucial role in electrical magneto-chiral anisotropy, demanding further in-depth investigation.

Influence of the Substrate on the Spatial Variation of Conductive Properties of Self-Assembled Monolayers

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Numerous studies observing chiral-induced spin selectivity (CISS) were carried out on self-assembled monolayers (SAMs) of molecules using conductive probe atomic force microscopy (CAFM) [1]. Especially in the early stages, this was done by measuring the spin-polarization of the current running through the SAMs on Au, which were metal layers deposited on Si/SiO$_2$ substrates, using magnetic probes [2].

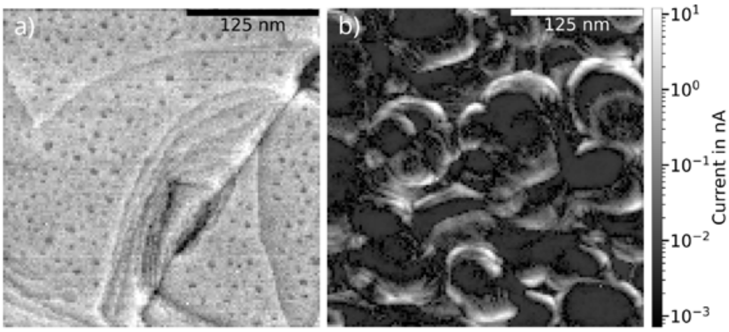

Figure 1 Current maps on mica/Au/DDT and Si/SiO$_2$/Au/DDT. The two maps present significant differences in the conductive properties of the different types of substrates.

Examining the importance of the choice of substrate on the lateral variation of the conductive properties of SAMs, we study one of the most basic molecules, dodecanethiol (DDT) and compare Si/SiO$_2$ to mica as the substrate for the Au layers. The studies show that the flatness of the substrate is of crucial importance for the spatial variation of the SAM's conductance. SAMs deposited on flat mica/Au show comparably homogeneous conductance spreading over large areas, while Si/SiO2/Au leads to strong variations with only small parts of the SAMs being comparable (see Fig. 1).

We conclude that the choice of substrate is essential for such studies and that for comparability purposes, a study of the spatial variation of the conductive properties of the SAM in question is favorable. This should hold for studies on alkanethiols, as well as other molecules that form SAMs.

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Influence of Nonequilibrium Vibrational Dynamics on Spin Selectivity in Chiral Molecular Junctions

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We employ a mixed quantum-classical approach combining Ehrenfest dynamics with hierarchical equations of motion (HEOM) to study the role of nonequilibrium vibrational dynamics in the chirality-induced spin selectivity effect in chiral molecule junctions. The chiral molecule junction consists of a chiral molecule sandwiched between two leads, one of them magnetic, while the other nonmagnetic. A previously introduced model Hamiltonian of the chiral molecule is used, which takes into account spin-orbit-vibrational couplings [2]. Within this approach, the dynamics of conduction electrons is treated fully quantum mechanically, whereas the dynamics of molecular vibrations is treated classically within the Ehrenfest approximation. We introduce a new figure of merit, the displacement polarization and show that this quantity is temporally correlated with the spin selectivity, indicating that vibrational dynamics contribute to a finite spin selectivity in this model. Averaging over an ensemble of trajectories, however, leads to a vanishingly small spin selectivity, which is attributed to a cancellation of single trajectory contributions. We analyze the ensemble distribution of spin selectivity and the temperature dependence, finding a temperature-dependent broadening of the distribution.

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Spin Hall Magnetoresistance as a Probe for Chiral-Induced Spin Selectivity

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The chiral-induced spin selectivity effect (CISS) describes the dependence of an electron's transmissivity through a chiral molecule on the molecule's chirality and the electron's spin. Despite numerous experimental and theoretical approaches, the fundamental mechanisms of the CISS effect are still an open question. A recent experiment [1] investigated the effect of chiral molecules on spin-to-charge conversion in a spin-pumping experiment to probe possible underlying phenomena of CISS. Based on this result, we will use a well-established experiment–the measurement of spin Hall magnetoresistance–that relies on the reflection and absorption of spin currents at interfaces adjacent to a layer that exhibits an (inverse) spin Hall effect. Combined with the results established in the spin-pumping experiment, this paves the path toward designing new chiral-molecule-based spin-torque devices.

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Towards Understanding First-Principles Electron–Phonon Coupling in CISS

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Considering charge transport phenomena, a connection of the electronic spin and the chirality of the conducting medium can be observed. This leads to a preferred spin state of the electron in e. g. molecular conduction of helical molecules, and is known as the chiral induced spin selectivity (CISS) effect [1]. Although the underlying mechanism is not yet fully understood, some experiments show an increase in spin polarization with increasing temperature, indicating a contribution based on molecular vibrations [2, 3]. Their effect on molecular conduction can be e. g. studied via inelastic electron tunneling spectroscopy (IETS), and an approach of Troisi *et al.* [4] in combination with relativistic two-component density functional theory was used to simulate spin-resolved IETS intensities.

Using a carbon-based model helix, helicity-dependent IETS spin polarizations are visible with polarizations up to ten times higher compared to spin polarizations emerging from elastic tunneling, even though at lower transmission intensities. Both inelastic and elastic tunneling spin polarizations show a strong dependency on the imaginary part of the molecular effective single-particle Hamiltonian matrix, which is furthermore strongly influenced by the chosen gold electrodes. First indications of correlations of vibrational properties (like the vibrational frequency or the resulting change in chirality) and spin polarizations could be observed. However, further steps towards more realistic systems and the calculation of a wider range of vibrational frequencies is required for a more profound understanding.

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Chirality-Dependent Electron Tunneling in the Small Molecule Limit

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In this work, ambient scanning tunneling spectroscopy (STS) with functionalized tips is modified towards a tunneling magnetoresistance setup. Using a systematic and modular approach with respect to the employed molecules, we paint a much less simplistic picture of CISS and the current spin polarization *SP* in the limit of small molecules (less than 20 atoms) than what appears as current consensus. Specifically, we show that (a) achiral molecule-substrate interactions can play a significant role in CISS, and (b) prolonged measuring of such junctions may lead to a structural or topological reorganization of the functionalized STS tip. Our results show that one or more effects – which are not necessarily chirality-dependent – compete with the CISS effect and that thus, *SP* is a poor predictor for the strength of CISS in the small-molecule limit.

An Electronic Spin on Nuclear Dynamics near Conical Intersections

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Molecules are inherently geometric in nature, as their properties are not only determined by their atomic composition but also their three-dimensional arrangement in space. However, the geometry arising from the atomic configuration alone is rather mundane, essentially being given by a tuple of Cartesian coordinates. A more exotic type of geometry emerges from the interplay between the nuclear coordinates and the electronic wave function. It is known, for instance, that the electronic wave function may not return to its initial state when transported around a closed contour in nuclear coordinate space, with the difference given by a phase factor.[1] This phase factor is geometric in nature only depending on the path taken in nuclear coordinate space and bearing some resemblances to the notion of curvature in Riemannian manifolds. When the nuclear motion is treated quantum mechanically according to the Born–Oppenheimer approximation, the geometric phase translates to an effective vector potential describing a pseudo-magnetic interaction with the nuclei. It can be argued that interactions of this type are most relevant if the electronic wave function is complex-valued, which, for instance, occurs if there is a conical intersection or if spin-orbit coupling is included.[2] Using a simple triatomic toy-model that exhibits a conical intersection at equilateral configuration, I will illustrate why geometric phase effects necessarily occur under these conditions. In addition, the results presented in this talk will be formulated using fibre bundles, providing a mathematical yet approachable perspective to the geometric phase effect.

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On the Origin of The Isotope Effects in Life

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Isotope effects in biomolecules may provide valuable insights into biochemical processes. By examining isotopic signatures in amino acids, we can determine if there were formed through abiogenic pathways or biological processes[1]. Previous research suggests that nuclear spin may influence isotopic effects in biological processes[2], hinting at a possible connection between spin-selective processes and isotope patterns in prebiotic or biotic chemistry. In our experiments, we crystallized L-Glutamic acid on magnetic surfaces and measured the isotopic ratios using mass-spectrometry. Preliminary findings indicate a time-dependent C-13 depletion at the magnet's north pole, which diminishes with longer crystallization times. These results link isotope effects to nuclear spin and spin selective processes, offering a new perspective to the earliest biochemical processes on Earth.

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Categories of Chirality: Full classification of enantiomorphism

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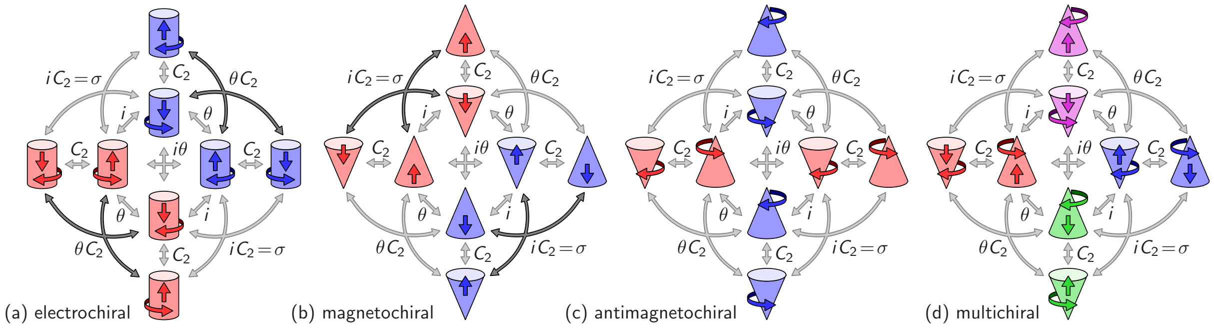
One speaks of chirality when a system exists in two versions (enantiomorphs) that cannot be superposed upon each other by proper (pure) rotations C [1]. Ordinarily, this is tantamount to the case that the enantiomorphs are mirror images of each other, i.e., the enantiomorphs are mapped onto each other by improper rotations iC, where i denotes space inversion. By treating space inversion i, time inversion θ, and their combination $i\theta$ on the same footing, we greatly expand the notion of chirality [2]. Besides the usual kind, which we call electrochirality because it arises from the interplay of electric multipolar order [2,3], we identify magnetochirality and antimagnetochirality as new categories of chirality characterized by the existence of two distinct enantiomorphs. While the enantiomorphs of electrochiral systems are mapped onto each other by improper rotations iC and $i\theta C$ but not θC [Fig. 1(a)], the enantiomorphs of magnetochiral systems are mapped onto each other by rotations θC and $i\theta C$, but not θC [Fig. 1(b)]. Similarly, the enantiomorphs of antimagnetochiral systems are mapped onto each other by rotations θC and iC, but not $i\theta C$ [Fig. 1(c)]. Multichiral systems also exist, where improper rotations iC, θC and $i\theta C$ each map the system onto a distinct enantiomorph, yielding four distinct enantiomorphs [Fig. 1(d)]. Finally, parachiral systems can always be mapped onto each other by improper rotations iC, θC and $i\theta C$, i.e., they do not support distinct enantiomorphs. The five categories of chirality identified here yield a complete classification of enantiomorphism, where any physical system belongs to exactly one category. We discuss implications of our classification relevant, in particular, for magnetically ordered systems.



(a) electrochiral (b) magnetochiral (c) antimagnetochiral (d) multichiral

Figure 1: Categories of chirality illustrated [2]. The transformation behavior under generalized improper rotations reveals enantiomorphism of simple objects commonly used to discuss chirality [1]. Straight (curved) arrows indicate translational (rotational) motion of an object.

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Chiral Molecular Coating of $LiNiCoMnO_2$ Cathode for High-Rate Capability Lithium-ion Batteries

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The growing energy demands increase the need for battery storage, with lithium-ion batteries being widely used. Among those, Nickel-rich layered lithium transition metal oxides ($LiNi_{1-x-y}Co_x Mn_yO_2$ - NCM) are one of the promising cathode materials due to their high specific capacities and working voltage. In this study, we demonstrated that a tmple layer coating of polyalanine chiral molecules improves the performance of Ni-rich cathodes. The chiral organic coating of the active material enhances the discharge capacity and the rate capability. Specifically, NCM811 and NCM622 electrodes coated with chiral molecules exhibit lower voltage hysteresis and better rate performance, with a capacity improvement larger than 10% at a 4C discharge rate and an average improvement of 6%. We relate these results to the chiral-induced spin selectivity (CISS) effect that enables to reduce the resistance of the electrode interface and reduce dramatically the overpotential needed for the chemical process by aligning the electrons spins.