MA 34: Molecular Magnetism

Time: Thursday 9:30–12:45 Location: H18

MA 34.1 Thu 9:30 H18

Handling higher order ligand field parameters of single molecule magnets using deep learning — •Zayan Ahsan Ali, Julius Mutschler, Preeti Tewatia, and Oliver Waldmann — Physikalisches Institut, Universität Freiburg, D-79104 Freiburg, Germany

In recent decades, Single Molecule Magnets (SMMs) have sparked an interest not only due to their applications in quantum computing and spintronics, but also as an ideal platform for exploring fundamental principles of quantum magnetism. While substantial progress has been made towards the characterization of magnetic properties of 3d SMMs, the study of 4f SMMs remains challenging. This difficulty arises from the involvement of up to 27 ligand field parameters and the typically featureless nature of experimental magnetic data, leading to severe overparameterization. Moreover, the physically relevant regions in this parameter space are mostly unknown a priori. Although deep learning based inverse models, such as Conditional Variational Autoencoders and Invertible Neural Networks, have shown promise in addressing overparameterization, their performance degrades significantly when trained on uninformative parameter spaces, which dominate especially in high dimensional settings. This work investigates the use of Monte Carlo based parameter sampling for the higher order ligand field parameter space as a crucial precursor towards improving the deep learning inverse models. The resulting dataset represents a more informative prior, enabling insights into the effects of higher order ligand field parameters and the correlations between them.

MA 34.2 Thu 9:45 H18

Polarization of Electron Spin and Orbitals in Chiral Molecular Junctions on Semiconductors — •Peng Xiong[1], Yuwaraj Adhikari[1], Tianhan Liu[1,3], Hailong Wang[2], Zhenqi Hua[1], Haoyang Liu[1], Paul Weiss[3], Binghai Yan[4], and Jianhua Zhao[2] — [1]Florida State University, USA — [2]Institute of Semiconductors, Chinese Academy of Sciences, China — [3]University of California, Los Angeles, USA — [4]Weizmann Institute of Science, Israel

Electrical generation and transduction of polarized electron spins in semiconductors via nonmagnetic means are of broad interest in spintronics and quantum information science. One such pathway is chirality-induced spin selectivity (CISS), where real-space structural chirality induces spin polarization of electrons from a nonmagnetic electrode. We have studied the CISS effect through measurement of spin-selective transport in chiral molecular junctions comprising a nonmagnetic normal metal electrode and a self-assembled monolayer of chiral molecules on magnetic (GaMnAs) or nonmagnetic (n-GaAs) semiconductors, where the spin polarization is detected via the spin-valve conductance and Hanle effect, respectively. The results reveal several important characteristics of the CISS effect [1-3]: i) nontrivial linear-response magnetoconductance in two-terminal CISS spin valves, in apparent violation of the Onsager reciprocal relation; ii) crucial role of the spin-orbit coupling in the normal metal electrode, suggesting the importance of orbital polarization in the chiral molecules; iii) spin generation by CISS in semiconductors. 1. ACS Nano 14, 15983 (2020); 2. Nat. Commun. 14:5163 (2023) ; 3. Ad. Mater. 36, 2406347 (2024).

MA 34.3 Thu 10:00 H18

Effects of Boundary Condition on Quantization in the Spin-1/2 Heisenberg Chain — •Saketh Ravuri[1], Chenxiao Zhao[1], Pascal Ruffieux[1], and Roman Fasel[1,2] — [1]Empa, Dübendorf, Switzerland — [2]University of Bern, Bern, Switzerland

The spin-1/2 antiferromagnetic Heisenberg chain resides in a gapless spin liquid phase in the thermodynamic limit. In finite-length systems, however, quantization introduces a length-dependent excitation gap, which is further influenced by the boundary conditions. In this work, we investigate how open and periodic boundary conditions affect the quantization and gap behavior in spin-1/2 antiferromagnetic Heisenberg chains constructed by covalently linking magnetic nanographene units. For chains of fixed length, we demonstrate that open boundary conditions result in a smaller gap compared to periodic boundary conditions, as clearly evidenced by the inelastic electron tunneling spectra. This impact of boundary conditions diminishes with increasing chain length and vanishes in the thermodynamic limit. Moreover, in periodic rings with odd-numbered units, we investigated the scattering of a single spinon caused by J-fluctuations. These findings illuminate the role of boundary effects in finite-size quantum spin systems and contribute to the fundamental understanding of quantum magnetism and excitations in spin chains.

MA 34.4 Thu 10:15 H18

Approximate finite-temperature Lanczos modelling of dysprosium containing magnetic molecules — •Jürgen Schnack and Dennis Westerbeck — Bielefeld University, Faculty of Physics, 33615 Bielefeld

Dysprosium containing magnetic molecules are considered promissing building blocks of future quantum technology such as storage, quantum computing, quantum sensing, or magnetocalorics. The initial physical characterization includes measurements of magnetization or heat capacity as function of temperature and applied magnetic field.

A theoretical modelling of an approximate quantum spin model by means of exact diagonalization of multicenter systems is virtually impossible due to the large magnetic moment of dysprosium. We resort to the finite temperature Lanczos method which however converges rather slowly [1]. We explain the reasons, our solutions and results for recent Dy containing molecules [2].

[1] O. Hanebaum and J. Schnack, Eur. Phys. J. B, 87, 194, 2014.

[2] D. Westerbeck, Ph.D. thesis, Bielefeld University, 2025, in preparation.

MA 34.5 Thu 10:30 H18

Annealing Induced Ordered Structures of H2Pc Monolayer on γ'-Fe4N Thin Film — •Hiroki Ono[1], Yoshitaka Umeda[1], Kaito Yoshida[1], Kenzaburo Tsutsui[1], Kohei Yamamoto[2], Osamu Ishiyama[2], Toshihiko Yokoyama[2], Masaki Mizuguchi[1], and Toshio Miyamachi[1] — [1]Nagoya University, Nagoya, Japan — [2]Institute for Molecular Science, Okazaki, Japan

Organic-inorganic hybrid interface has been studied because its interfacial spin state can be controlled through proximity effect. Spin state depends on local interface structures as well as electronic interaction between organic molecules and magnetic substrates. Therefore, controlling the interface structure is essential but remains challenging because of strong interaction between organic and inorganic materials.

In this work, we fabricate organic-inorganic hybrid thin films using iron nitride as a ferromagnetic substrate and phthalocyanine(H2Pc) as an organic molecule. Iron nitride atomic layers with γ'-Fe4N stoichiometry uniformly grow and show weaker interaction than typical ferromagnetic substrate. Therefore, γ'-Fe4N is a candidate to create structurally controlled interface. We investigate interface structure by using scanning tunneling microscopy(STM) and low energy electron diffraction(LEED), and electronic/magnetic properties by using x-ray absorption spectroscopy/magnetic circular dichroism(XAS/XMCD). We confirm the magnetic coupling occurs at H2Pc/γ'-Fe4N bilayer interface from XAS/XMCD results. H2Pc monolayer on γ'-Fe4N bilayer do not have any long-range lattice manner. But, we find annealing induces well-ordered structure while preserving interface magnetic coupling.

MA 34.6 Thu 10:45 H18

CuCu4 metallacrowns on the Au(111) surface: A density functional study — •Ariyan Tavakoli[1], Stefan Lach[1], Benjamin Stadtmüller[2], Christiane Ziegler[1], Peter Puschnig[3], and Hans Christian Schneider[1] — [1]Department of Physics and Research Center OPTIMAS, University of Kaiserslautern-Landau, Kaiserslautern, Germany — [2]Experimentalphysik II, Institute of Physics, Augsburg University, Augsburg, Germany — [3]Department of Physics, University of Graz, Graz, Austria

Metallacrowns [1] combine chemical and structural features that make them a promising material system for single-molecule magnets. Here, we present a first-principles study of the electronic and magnetic properties of the metallacrown (HNEt3)2CuII [12-MCYN(Shi)-4] (Y=CuII), in short CuCu4, in the gas phase and on the Au(111) surface. First, we study the magnetic properties of CuCu4 metallacrown in the gas phase by applying the broken symmetry approach [2], where we benchmark the performance of various (range-separated) hybrid functionals compared to the computationally cheaper GGA+U approach. In the second step, we explore the magnetic configurations of CuCu4 metallacrown adsorbed on an Au(111) surface using density functional theory (DFT) with GGA+U framework. The analysis highlights the changes of the ligand structure and the density of state (DOS) around the metal centers by a comparison between the isolated molecule and the adsorbed one on the surface.

[1] B. R. Gibney et al., Inorganic Chemistry 33 (1994). [2] Pavlyukh, Y. et al., PhysRevB. 99, 144418 (2019).

15 min. break

MA 34.7 Thu 11:15 H18

Cooperative and Selective Redox Doping Switches Single-Molecule Magnetism — Fabian Paschke[1], Matteo Briganti[2], Vivien Enenkel[1], Tobias Birk[1], Jan Dreiser[3], Peter Schmitt[4], Rainer F. Winter[4], Federico Totti[2], and •Mikhail Fonin[1] — [1]Fachbereich Physik, Universität Konstanz, 78457 Konstanz, Germany — [2]Department of Chemistry 'Ugo Schiff' and INSTM Research Unit, University of Florence, 50019 Sesto Fiorentino, Italy — [3]Swiss Light Source, Paul Scherrer Institute, 5232 Villigen PSI, Switzerland — [4]Fachbereich Chemie, Universität Konstanz, 78457 Konstanz, Germany

The controlled manipulation of electronic and magnetic states in single-molecule magnets (SMMs) is crucial for their implementation in molecular spintronics. In typical SMMs, key properties like magnetic anisotropy and slow magnetic relaxation are imposed by complex ligand shells, whose bulky and three-dimensional structures hamper efficient manipulation of the molecular magnetism by chemical methods. Here, we demonstrate highly selective chemical doping of an Fe_4 nanomagnet on a Pb(111) surface using lithium atoms. Scanning tunneling microscopy, X-ray absorption spectroscopy, and ab initio calculations reveal the cooperative incorporation of three Li atoms per Fe_4 molecule, resulting in a selective, threefold reduction of its iron-based magnetic core. The doping modifies the intramolecular exchange interaction, turning from antiferromagnetic to ferromagnetic coupling, and changes the molecular magnetic anisotropy from easy-axis to easy-plane.

MA 34.8 Thu 11:30 H18

Tuning spin-injection into metallacrown/thin-metal film systems — •David Anthofer[1], Ashish Moharana[1], Dominik Laible[2], Fabian Kammerbauer[1], Alexander Hagenow[2], Eva Rentschler[2], and Angela Wittmann[1] — [1]Institut für Physik, Johannes-Gutenberg Universität Mainz, Deutschland — [2]Department Chemie, Johannes-Gutenberg Universität Mainz, Deutschland

Single-molecular magnets (SMMs) have recently gained significant interest due to their ability to retain magnetic information at the molecular level, offering potential applications in high-density data storage devices. A crucial challenge hindering their application in technology is the integration with thin-film devices. To tackle this challenge, we explore the spin-injection efficiency in hybrid molecule/non-magnetic metal thin film heterostructures to understand the impact of hybridization. For this, we utilize molecules based on the metallacrown system, chosen for their unique combination of synthetic versatility and structural stability. We inject a pure spin current at ferromagnetic resonance into the hybrid interface, allowing us to measure the magnetic damping of the system. We observe a notable increase in damping after adsorption of Dysprosium-based metallacrown SMMs. In contrast, no change in damping was observed after deposition of Copper-based metallacrowns indicating the significance of molecular composition on the spin-injection efficiency at the hybrid interface. Further optimization of the ferro- and nonmagnetic metal layer thicknesses proves to affect the observed change in damping, paving the path toward a sensitive framework to study hybridization at the molecule/metal interface.

MA 34.9 Thu 11:45 H18

Probing magnetic exchange interactions in cobalt-based molecular magnets using magneto-Raman spectroscopy — •Komalavalli Thirunavukkuarasu[1], David Hunger[2], Julia Netz[3], Duncan Moseley[4], Ziling Xue[4], Dmitry Smirnov[5], Andreas Koehn[3], and Joris van Slagaren[2] — [1]Department of Physics, Florida A & M University, Tallahassee, FL, USA — [2]Institute of Physical Chemistry, University of Stuttgart, Stuttgart, Germany — [3]Institute of Theoretical Chemistry, University of Stuttgart, Stuttgart, Germany — [4]Department of Chemistry, University of Tennessee, Knoxville, USA — [5]National High Magnetic Field Laboratory, Tallahassee, USA

Combining spectroscopy with one or more external parameters such as low temperature, high pressure, and high magnetic fields, allows us to probe interplay between spin, charge, orbital, and lattice degrees of freedom. Spin exchange interactions play an important role in single-molecule magnets and molecular qubits that feature magnetic bistability. However, there have been few detailed studies to experimentally find the energy scale of these interactions such as spin-phonon coupling. Recently, we employed magneto-Raman spectroscopy together with theoretical work on single molecular magnets to reveal the signatures of spin-phonon interactions in these materials. In this talk, the magneto-Raman experimental results from mononuclear cobalt(II) and radical-bridged dinuclear cobalt(II) complexes and the outcome will be discussed.

MA 34.10 Thu 12:00 H18

The Role of Quantum Vibronic Effects in the Spin Polarization of Charge Transport through Chiral Molecular Junctions — •Samuel Rudge[1], Christoph Kaspar[1], Riley Preston[1], Joseph Subotnik[2], and Michael Thoss[1] — [1]Institute of Physics, University of Freiburg — [2]Department of Chemistry, Princeton University

The chirality-induced spin selectivity (CISS) refers to the experimentally observed phenomenon that the transport of spin-polarized electrons through chiral mediums can be highly asymmetric between the two spin orientations and enantiomers [1]. Although the exact mechanism underpinning the CISS effect is still unknown, one of the leading ideas is that it is connected to the coupling of transport electrons to molecular vibrations. In this contribution, 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 [2]. Specifically, we calculate charge currents through a two-site, two-mode model [3], focusing on the highly nonadiabatic regime of low-voltage charge transport, in which we find significant spin polarization.

[1] R. Naaman and D. H. Waldeck, Annu. Rev. Phys. Chem. **66**, 263-281 (2015)

[2] C. Schinabeck, A. Erpenbeck, R. Härtle2, and M. Thoss, Phys. Rev. B **94**, 201407(R) (2016)

[3] H.-H. Teh, W. Dou, and J. Subotnik, Phys. Rev. B **106**, 184302 (2022)

MA 34.11 Thu 12:15 H18

Chiral-induced unidirectional spin-to-charge conversion — •Ashish Moharana[1], Yael Kapon[2,3], Fabian Kammerbauer[1], David Anthofer[1], Shira Yochelis[2,3], Mathias Kläui[1], Yossi Paltiel[2,3], and Angela Wittmann[1] — [1]Institute of Physics, Johannes Gutenberg University Mainz, Mainz 55128, Germany — [2]Institute of Applied Physics, The Hebrew University of Jerusalem, Jerusalem 9190401, Israel — [3]Center for Nanoscience and Nanotechnology, The Hebrew University of Jerusalem, Jerusalem 9190401, Israel

The chiral-induced spin selectivity (CISS) effect has recently gained significant attention in the field of spintronics. 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. While so far research has predominantly focused on transport properties, in our work, we explore spintronic phenomena at hybrid chiral molecule magnetic interfaces to elucidate the underlying mechanisms of the chiral-induced spin selectivity effect. For this, we investigate the interfacial spin-orbit coupling in chiral molecule/metal thin film heterostructures by probing the chirality and spin-dependent spin-to-charge conversion. Our findings validate the central role of spin angular momentum for the CISS effect, paving the path toward the functionalization of hybrid molecule-metal interfaces via chirality.

MA 34.12 Thu 12:30 H18

Interplay between spin induced polarization and quantum entanglement in triangular magnetic molecules — •Zhirayr Adamyan[1,2], Vadim Ohanyan[1,2], and Ani Chobanyan[1] — [1]Laboratory of Theoretical Physics, Yerevan State University, 1 Alex Manoogian, 0025 Yerevan, Armenia — [2]CANDLE, Synchrotron Research Institute, 31 Acharyan Str., 0040 Yerevan, Armenia

The quantum entanglement of spin states in molecular magnets has important applications in quantum information technologies and quantum computing. Currently, qubit models based on magnetic molecules are being used to develop quantum computation and communication technologies. We consider two models of three-spin molecular magnets with additional features that allow one to manipulate and enhance their entanglement. The first model is a mixed-spin (1/2, 1, 1/2) triangle with two g-factors. The second model is a spin-1/2 triangle with the Katsura-Nagaosa-Balatsky (KNB) mechanism, providing the coupling between spin degrees of freedom and the external electric field. It is shown that non-conserving magnetization originated from the non-uniformity of g-factors leads to an essential increase of the entanglement of certain spin states along with the rich structure of zero-temperature phase diagrams. Whereas, the model with magneto-electric coupling due to the KNB mechanism offers a wide possibility of manipulation of quantum entanglement by the electric field, both using its magnitude and direction.