MA 36: Molecular Magnetism and Magnetic Particles / Clusters I

Time: Wednesday 15:00–18:30 Location: POT/0151

MA 36.1 Wed 15:00 POT/0151

A Combined Theoretical and Experimental Study of Oxygen Vacancies in Co_3O_4 for Liquid-Phase Oxidation Catalysis — Amir Omranpour[1], •Lea Kämmerer[2], Catalina Leiva–Leroy[1], Anna Rabe[2], Takuma Sato[3], Soma Salamon[2], Joachim Landers[2], Benedikt Eggert[2], Eugen Weschke[4], Jean Pascal Fandré[5], Ashwani Kumar[5], Harun Tüysüz[5,6], Martin Muhler[1], Heiko Wende[2], and Jörg Behler[1] — [1]University of Duisburg-Essen and CENIDE — [2]Ruhr-University Bochum — [3]Max Planck Institute for Chemical Energy Conversion — [4]Helmholtz-Zentrum Berlin für Materialien und Energie — [5]Max-Planck-Institut für Kohlenforschung — [6]IMDEA Materials Institute, Spain

We combined Density Functional Theory (DFT) and experimental techniques to investigate oxygen vacancies (V_O) in bulk Co_3O_4 and during liquid-phase ethylene glycol oxidation. DFT calculations show that a V_O reduces two adjacent Co^{3+} ions to stable, high-spin Co^{2+} in distorted octahedral sites, simultaneously narrowing the band gap. Comparison of experimental O K-edge X-ray absorption spectra with DFT calculated spectra reveal that the fresh catalyst resembles the vacancy-containing calculation, but the post-reaction catalyst shifts toward the ideal Co^{3+} state, strongly suggesting that Co_3O_4 becomes more oxidized under liquid-phase ethylene glycol oxidation by refilling preexisting oxygen vacancies, a finding supported by increased conversion at higher O_2 pressures and the catalyst's stability and activity over multiple cycles. We gratefully acknowledge the DFG funding by CRC/TRR 247 (Project ID: 388390466) projects A01, A10, and B02.

MA 36.2 Wed 15:15 POT/0151

KNB mechanism in convex polygon molecular magnets: bipartite entanglement transfer with the aid of electric field. — •Zhirayr Adamyan[1,2], Vadim Ohanyan[1,2], Ani Chobanyan[1], Hamid Arian Zad[3], Jozef Strecka[3], Azadeh Ghannadan[4], and Saeed Haddadi[4,5] — [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 — [3]Department of Theoretical Physics and Astrophysics, Faculty of Science, P. J. Safarik University, Park Angelinum 9, 041 54 Kosice, Slovak Republic — [4]Saeeds Quantum Information Group, P.O. Box 19395-0560, Tehran, Iran — [5]Faculty of Physics, Semnan University, P.O. Box 35195-363, Semnan, Iran

The unique properties exhibited by single molecular magnets (SMMs) have led to their integration into hybrid devices, emphasizing their quantum nature. We examine models of 1/2-spin molecular magnets arranged in a convex polygon configuration, utilizing a Katsura-Nagaosa-Balatsky (KNB) mechanism to couple the spin degrees of freedom to an external electric field. This KNB mechanism enables extensive control over quantum entanglement through both the magnitude and direction of the electric field. By employing a rotating configuration of the KNB-coupled electric field, where the field's magnitude remains constant while its direction changes, we demonstrate the controllable transfer of bipartite entanglement between different pairs of spins in the model.

MA 36.3 Wed 15:30 POT/0151

Magneto-optical properties of ferromagnetic liquid crystals — •Tom Rehfeldt[1], A. Jarosik[1], D. Lisjak[2], A. Eremin[1], and H. Nádasi[1] — [1]Institute of Physics, Otto von Guericke University, Universitätsplatz 2, 39106 Magdeburg, Germany — [2]Jožef Stefan Institute, Jamova cesta 39, Ljubljana SL-1000, Slovenia

Ferromagnetic nematic liquid crystals (FNLCs) combine the orientational order of nematic liquid crystals with the spontaneous magnetization of ferrimagnetic nanoplatelets.

In this work, the dynamic magneto-optical behavior of FNLCs is investigated by changing the parameters of the external dynamic magnetic field.

MA 36.4 Wed 15:45 POT/0151

Making molecular changes visible using near-surface transport of micron-sized magnetic particles — •Yahya Shubbak, Nikolai Weidt, Arne Vereijken, Rico Huhnstock, and Arno Ehresmann — Institute of Physics and Center for Interdisciplinary Nanostructure Science and Technology (CINSaT), University of Kassel, Heinrich-Plett-Strasse 40, 34132 Kassel, Germany

Transport of magnetic particles (MPs) in liquid close to the surface of a flat substrate containing a periodic magnetic domain pattern is a promising lab-on-a-chip (LOC) technology for detecting MP-bound analytes, even when their size is negligible compared to the MP size. As proof of principle, we show that simple observation of MP motion via optical microscopy is sufficient to distinguish MPs of the same nominal size, but surface-functionalized with two different functional groups. The different surface chemistry changes the liquid-mediated MP-to-substrate forces acting during close-to-surface transport, resulting in significant variations in the experimentally observable MP velocity. More specifically, superparamagnetic MPs measuring 2 micrometre in diameter with a polymer coating of solely carboxyl (COOH) end groups, or a mixture of carboxyl and amino (NH2) groups, respectively, have been studied. Transport of these MPs above a magnetic stripe domain pattern in double-distilled water showed a remarkable difference in their average velocities, rendering the COOH-functionalized MPs almost twice as fast as the NH2 counterparts for otherwise identical experimental parameters. This result enables the magnetophoretic separation of MPs based on their surface properties.

MA 36.5 Wed 16:00 POT/0151

Magnetic cuboidal particles as field sensors for microscaled magnetic stray field landscapes — •Jonas Bugase, Christian Janzen, Arne Vereijken, Yahya Shubbak, Nikolai Weidt, Rico Huhnstock, and Arno Ehresmann — Institute of Physics , University of Kassel, 34132 Kassel

Recently, magnetic domain patterns engineered in thin-film systems have been widely used in data storage, spintronics, and magnetic imaging. Characterizing the micron-scale magnetic stray fields emerging from these patterns remains a topical but challenging task [1]. We present a non-mechanical, optically trackable method to visualize effective magnetic field directions using anisotropically shaped magnetic particles. These particles are fabricated via 2-photon polymerization lithography and magnetic exchange-biased layer deposition to achieve custom shapes and anisotropies. When positioned in the field landscape of a periodically patterned substrate [2] within a quiescent liquid, the particles spontaneously orient and reorient in response to local micron-scale stray fields. This enables efficient characterization of complex magnetic domain patterns. We further explore probing local field variations with superimposed external fields facilitating diagnostic and biomedical microfluidic applications [3].

[1] Nistico *et al.* (2020), Inorganics, 8(1):6.
[2] Holzinger *et al.* (2013), J. Appl. Phys, (114): 013908.
[3] Ehresmann *et al.* (2015), Sensors, (15): 28854.

MA 36.6 Wed 16:15 POT/0151

Metal-free magnetism and organic 2D crystals — •Hongde Yu and Thomas Heine — TU Dresden, Dresden, Germany

Organic 2D crystals with metal-free magnetism have attracted considerable research interest owing to their promising applications in organic spintronics and quantum information technologies. However, achieving stable spin-polarization and controlling magnetic interactions in these systems remains challenging due to strong electronic coupling and the closed-shell nature of most organic monomers. In this talk, I will present a strategy to induce spin-polarization and tailor magnetic interactions in organic 2D crystals. By assembling triangulene monomers into dimers and extended 2D polymers, we theoretically explore strategies to control magnetic interactions and electronic structures. We showed substantial magnetic coupling (J) up to -198 meV through rational chemical design of triangulene dimers. Furthermore, triangulene-based 2D crystals exhibit unique electronic features, including a Dirac point flanked by twin flat bands. By tuning the Fermi level, we predict metal-free ferromagnetism with Curie temperatures of ~260 K and half-metallic behavior. Furthermore, we proposed a mixed-topology design strategy enabling purely organic 2D FM semiconductors with J values up to 127 meV and Curie temperatures exceeding 500 K. Beyond conventional FM and AFM, I will also present a simple approach to achieve metal-free altermagnetism.

MA 36.7 Wed 16:30 POT/0151

Modelling the Spin States of Frustrated Nanographenes

and Their Dimers: Challenges & Insights — •Helen Preiss, Hongde Yu, and Thomas Heine — Dresden University of Technology, Germany

Metal-free magnetism is an emerging field with promising applications especially in quantum computing. Fundamentally, metal atoms are replaced by organic radicals as spin-carriers, which can be extended to 1D or 2D networks via suitable coupling reactions while possibly maintaining a long-range magnetic order. One class of such radical building blocks are the topologically frustrated nanographenes. For these, it is impossible to pair all π-electrons into alternating single and double bonds, resulting in an open-shell system. A prominent example is the recently synthesised Clar's goblet, a biradical with a singlet ground-state according to Lieb's theorem. We will show that BS-DFT fails to quantitatively predict the magnetic coupling in this molecule, and to describe its dimer even qualitatively, owing to the delocalised nature of the spin centres and their inherent multi-reference character, while higher level ab initio methods struggle with the sheer size of the systems. Alternatively, Clar's goblet may be thought of as two olympicenyl radicals fused together by a six-membered ring. Sublattice-balanced and C-C-linked olympicenyl dimers thus offer themselves as a comparable yet simpler system to study. By comparing both types of nanographene, we gain insights into the limitations of modelling the magnetic states and both inter- and intramolecular coupling of organic molecules with BS-DFT.

15 min break

MA 36.8 Wed 17:00 POT/0151

Tuning chiral-induced unidirectional spin-charge conversion via molecular organization — •Ashish Moharana[1], Zhitian Ling[2], Hao Wu[2], Tomasz Marszalek[2], and Angela Wittmann[1] — [1]Institut für Physik, Johannes-Gutenberg-Universität Mainz, 55099 Mainz, Germany — [2]Max Planck Institute for Polymer Research, Ackermannweg 10, 55128, 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 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 an open challenge. In our work, we explore spin-to-charge conversion at hybrid chiral helical nanographene and heavy metal interfaces. We demonstrate that spin-to-charge conversion in hybrid chiral heterostructures can be tuned by controlling the ordering, and crystallinity of helical nanographene molecules at the interface. We show a more than two-fold increase in interfacial CISS efficiency with increasing molecular crystallinity. Quantifying the impact of spin to charge as a function of the molecular structure reveals the role of the molecular design and organization in the spin filtering effect, paving the path toward the three-dimensional engineering of hybrid interfaces.

MA 36.9 Wed 17:15 POT/0151

Revealing molecular alignment in thin films of DyCu5 single-molecule magnets via XMCD — •David Anthofer, Ashish Moharana, Alexander Hagenow, Dominik Laible, Tristan Fischer, Eva Rentschler, Hans-Joachim Elmers, and Angela Wittmann — Johannes-Gutenberg Universität Mainz, Deutschland

Single-molecule magnets (SMMs) have recently gained significant interest due to their ability to retain magnetic information at the molecular level, offering potential applications in ultra-compact and high-density data storage devices. A crucial challenge hindering their application in technology is the integration with thin-film devices. To tackle this, we probe the magnetic properties of DyCu5 SMMs deposited by dip-coating on a gold surface, where layer thickness is controlled by rinsing or not rinsing the sample. Using X-ray magnetic circular dichroism combined with sum-rule analysis for 4f elements, we extract the spin and orbital moments of the central Dy3+ ion. Rotating the sample relative to the incident beam reveals magnetic anisotropy in the thin layer, indicating preferential molecular alignment at the interface. In thicker films, this anisotropy vanishes, suggesting that the molecules are ordered on top of the gold surface, but have a random orientation with increasing coverage. While engineering ordered molecular assemblies on surfaces is generally challenging, our results demonstrate that a readily accessible dip-coating protocol can induce ordered alignment in the low-coverage regime, providing a practical route toward incorporating SMMs into thin-film devices.

MA 36.10 Wed 17:30 POT/0151

Probing the magnetic behavior of the metastable high-spin state achieved by light-induced excited spin-state trapping in Fe (II) spin-crossover complexes — •Sangeeta Thakur[1], Marcel Walter[1], Tarek Al Said[2], Eike F. Kuhlemann[3], Clara W. A. Trommer[3], Torben Adam[3], Karsten Holldack[2], Felix Tuczek[3], and Wolfgang Kuch[1] — [1]Institut für Experimentalphysik, Freie Universität Berlin, Arnimallee 14, 14195 Berlin, Germany — [2]Helmholtz-Zentrum Berlin für Materialien und Energie, Hahn-Meitner-Platz 1, 14109 Berlin, Germany — [3]Institut für Anorganische Chemie, Christian-Albrechts Universität zu Kiel, 24098 Kiel, Germany

The magnetic behavior of the metastable high-spin (HS) state of the Fe (II) complexes mononuclear [Fe(bpz)$_2$(bipy)]; (bpz = dihydrobis(pyrazolyl)borate) and dinuclear [(Fe(bpz)$_2$)$_2\mu$-(ac(bipy)$_2$)] (ac(bipy)$_2$ = bridging ligand) was investigated at 5 K by frequency-domain Fourier transform electron paramagnetic resonance (EPR) spectroscopy in a magnetic field of 1-10 T. The magnetic field map of the EPR measurements indicates a large value (70-90 cm^{-1}) of zero-field splitting (ZFS) for the dinuclear and mononuclear complexes. Comparatively, the HS-locked complexes [Fe(H$_2$B(pz)$_2$)$_2$(4,7-Me$_2$-phen)] and [Fe(tris(3-Methylpyrazolyl)borate)$_2$] exhibit values of ZFS around around 10-17 cm^{-1}. The large ZFS obtained for the dinuclear and mononuclear complexes could be due to the contribution of spin-orbit coupling (SOC). To the best of our knowledge, this is the first time the spin Hamiltonian parameters for Fe (II) SCO complexes were investigated in the metastable HS state.

MA 36.11 Wed 17:45 POT/0151

Insights into magnetic and magnetocaloric features of 3d-4f nonanuclear Gd3Cu6 complex from a mixed spin-(7/2, 1/2) Heisenberg diamond cluster — •Jozef Strecka and Katarina Karlova — Pavol Jozef Safarik University, Kosice, Slovakia

Low-temperature magnetization curves and magnetocaloric features of 3d-4f molecular complex [Gd3Cu6L6(OH)6(CH3OH)6(H2O)6]Cl3.5H2O (LH2 = 1,1,1-trifluoro-7-hydroxy-4-methyl-5-aza-hept-3-en-2-one) to be further abbreviated as Gd3Cu6 are examined within the framework of exact diagonalization method as well as the cluster-based mean-field theory. The magnetic compound Gd3Cu6 reveals in low-temperature magnetization curves an intermediate magnetization plateau at 21.27 Bohr magnetons, which is far below the saturated magnetization value of 27 Bohr magnetons expected for three spin-7/2 Gd3+ and six spin-1/2 Cu2+ magnetic ions per magnetic molecule. The crystal structure of the Gd3Cu6 complex is built from three condensed cubane-like moeties Gd2Cu2, which are from the magnetic point of view equivalent with three corner-sharing diamond motifs. The nonanuclear coordination compound Gd3Cu6 thus affords an intriguing experimental realization of the mixed spin-(7/2, 1/2) Heisenberg diamond chain composed from three unit cell. The magnetic compound Gd3Cu6 displays at sufficiently low temperatures an enhanced magnetocaloric effect, which is due to a geometric spin frustration quite superior with respect to that one of paramagnetic salts built from Gd3+ magnetic ions.

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MA 36.12 Wed 18:00 POT/0151

Suppressing geometric frustration in triangular dysprosium cluster encapsulated in fullerenes — •Matheus Barbosa, Wei Yang, Noel Israel, Fupin Liu, Bernd Büchner, Stanislav Avdoshenko, and Alexey Popov — Leibniz Institute for Solid State and Materials Research, Dresden/Germany

Nitride clusterfullerenes are compounds in which a metallic cluster is encapsulated within a fullerene cage, exhibiting diverse magnetic properties that depend on the composition and geometry of the endohedral cluster. For the case of $Dy_3N@C_{80}$, the presence of the central nitrogen ion induces a strong uniaxial Ligand Field contribution, locking the magnetic moments of dysprosium centers in a triangular arrangement. Such geometry avoids simultaneous ferromagnetic alignment of the moments between the dysprosium ions, leading to ground state frustration and exhibiting rather efficient spin reorientation through the Quantum Tunneling channel. Their magnetic and geometric frustration is harmful for the Single Molecule Magnetism, resulting in closed hysteresis loops in zero-field and at 2 K. We here report the suppression of this frustration by functionalization of the fullerene cage. The photochemical reaction of $Dy_3N@C_{80}$ with adamantane aziridine produced the monoadduct $Dy_3N@C_{80}$-Ad. This additional space inside the fullerene

has direct influence on the cluster, elongating the atomic distance in the Dy-N bond towards the modified fragment (breaking the spatial symmetry) and substantially increasing the Dy*Dy coupling constants. As a result, the monoadduct $Dy_3N@C_{80}$-Ad exhibits suppression of the geometric frustration and enhanced Single Molecule Magnetism.

MA 36.13 Wed 18:15 POT/0151

Orientation of Magnetic Anisotropy Axes in Single Molecule Magnets by Inelastic Neutron Scattering: A Case Study on Mn_2Y_2 — •Yifan Wang[1], Christoper E. Anson[2], Zayan A. Ali[1], Annie K. Powell[2], and Oliver Waldmann[1] — [1]Physikalisches Institut, Universität Freiburg, Germany — [2]Institut of Inorganic Chemistry, Karlsruhe Institute of Technology (KIT), Germany

Single molecule magnets (SMMs) are molecular systems that exhibit slow magnetic relaxation and well-defined spin states arising from strong magnetic anisotropy. These features give them potential for future information storage and have attracted much research interest. However, determining the magnetic parameters experimentally, especially the anisotropy orientations, remains challenging. In this work, we demonstrate, with regard to this point, the capability of inelastic neutron scattering (INS) on powder samples through a case study on the dimeric molecular spin cluster Mn_2Y_2. This complex belongs to a family of SMMs and consists of two magnetic spin-2 Mn^{III} ions coupled by antiferromagnetic exchange. The recorded INS spectrum on Mn_2Y_2 displays three cold transitions corresponding to singlet-triplet excitations. The transition energies enable a precise determination of the exchange coupling and the anisotropy magnitudes of the Mn^{III} centers. Most interestingly, a detailed analysis of the peak intensities is shown to yield, in addition, information on the orientation of the Mn^{III} magnetic anisotropy axes. The approach can be generally extended to related SMM families and provides a valuable addition to the range of experimental techniques.