MA 6: Molecular Magnetism

Time: Monday 9:30–13:00 Location: EB 301

Invited Talk MA 6.1 Mon 9:30 EB 301

Studying single molecule magnets for quantum technologies — •Wolfgang Wernsdorfer — PHI and IQMT, KIT, Karlsruhe, Germany

Single molecule magnets (SMMs) have been proposed for applications in high-density storage, quantum sensing, quantum simulation, quantum computing, and spintronics applications. Bulk magnetometric and spectroscopic techniques of molecular systems have allowed the observation of remarkable quantum effects in SMMs, such as the observation of an energy barrier to the reversal of the magnetisation and quantum tunnelling of the magnetisation Over the past 10 years, scanning tunnelling microscopy of SMMs and single-molecule devices architectures, such as spin valves and spin transistors, have shed light into the quantum properties of SMMs at single molecule level. More recently, new techniques, where the spin-degrees of freedom in SMMs can be read-out by photons, are being studied. Here, we review key techniques allowing the observation of quantum effects, important for the initialisation, control and read-out of the states of the SMMs, ultimately leading to the implementation of SMMs is technological applications. In the long term, chemically designed quantum architectures might have the potential to outperform existing platforms in terms of scalability, switchability, controllability, qubit density, and integrability.

MA 6.2 Mon 10:00 EB 301

Study of Landau-Zener transition of electron spin state on Single Ho atom — •Wonjun Jang[1,2], Luciano Colazzo[1,2], Georg A. Traeger[4], Lei Fang[1,2], Fabio Donati[1,3], Chau Bui[1,3], Soo-Hyon Phark[1,3], and Andreas Heinrich[1,3] — [1]Center for Quantum Nanoscience, Institute for Basic Science, Seoul 03760, South Korea — [2]Ewha Womans University, Seoul 03760, South Korea. — [3]Department of Physics, Ewha Womans University, Seoul 03760, Republic of Korea — [4]Physik, Georg-August-Universität, Göttingen, Germany

We report Landua-zener transition of electron spin states of a single Holmium (Ho) atom using spin-polarized scanning tunneling microscopy (STM). Single Ho atom on an oxygen site on MgO results in avoided level crossings due to the crystal field and hyperfine interaction. These avoided level crossings manifest as nuclear spin-mediated Landau-Zener tunneling of the electron spin state. By employing a combination of magnetic field sweeping and spin-polarized scanning tunneling microscopy, we measured the probabilities of electron spin reversal through Landau-Zener tunneling at the avoided level crossing with an energy gap of 5 peV. The maximum probability observed is 16%, influenced by the thermal population of nuclear spin states. Our spin-polarized STM measurement at the specific avoided level crossing enabled the single-shot measurement of the nuclear spin state. This research represents a direct measurement of the time evolution of the nuclear spin state of a rare earth atom.

MA 6.3 Mon 10:15 EB 301

Real-Space Imaging of Triplon Excitations in Engineered Quantum Magnets — •Robert Drost[1], Shawulienu Kezilebieke[2], Jose Lado[1], and Peter Liljeroth[1] — [1]Aalto University, Department of Applied Physics — [2]University of Jyväskylä, Department of Physics, Department of Chemistry, and Nanoscience Center

Despite the absence of long-range order, quantum magnetic ground states result from the inter-actions of spins at the nanoscale. It is thus possible to rationally design quantum magnets with pre-defined properties from simple ingredients. Organo-metallic molecules provide highly flexible spin systems. This flexibility makes them ideal candidate building blocks for designer quantum magnets. One example of fundamental excitations in quantum magnets are triplons. These dispersive triplet modes result form the internal excitations in the building blocks. Here, we show that triplon excitations can be produced in designer quantum systems and probed in real space using scanning tunneling microscopy (STM). We achieve this using assemblies of metal phthalocyanine molecules with an internal singlet-triplet transition. We further show that the dispersion bandwidth of triplons is strongly correlated with the dimensionality of the molecular assembly as expected from dispersive many-body modes. Our experiments show that arrays of metal-organic molecules are efficient platforms to simulate quantum magnets and study their excitations in a simplified setting.

MA 6.4 Mon 10:30 EB 301

High-field/high-frequency electron paramagnetic resonance studies on a muffin-shaped Er(III) complex — •Birte Beier[1], Jan Arneth[1], Gerlinde Greif[2], Peter Roesky[2], and Rüdiger Klingeler[1] — [1]Kirchhoff Institute for Physics, Heidelberg University, Germany — [2]Institute for inorganic chemistry, Karlsruhe Institute for Technology, Germany

Quantitative determination of magnetic anisotropy and the energy level diagram in $4f$ monomeric complexes is key to design appropriate crystalline environments of the magnetic centers. Here, we report high-field/high-frequency electron paramagnetic resonance spectroscopy (HF-EPR) and magnetisation studies on the phenantrolin-pyridin-triazol (PPT) complex $[Er(PPTMP)_2(MeOH)][OTf]_3$x3MeOH which is characterised by a muffin-shaped first coordination sphere. Our observation of a Kramer's doublet with $g_{\text{eff}} = 8.5(4)$ implies a $m = \frac{7}{2}$ ground state doublet of the 9-fold coordinated Er(III) ion. This conclusion is further supported by high-field magnetisation data. The absence of inter doublet transitions indicates that the gap to the first excited Kramers doublet exceeds 550 GHz. The anisotropic effective g-factors are compared with numerical studies and discussed with respect to related Er(III) complexes.

MA 6.5 Mon 10:45 EB 301

Room Temperature Ferromagnetism in $Tb_3N@C_{80}$ Crystals — •Lebin Yu[1], Shangfeng Yang[2], and Thomas Greber[1] — [1]Physik-Institut, University of Zürich, Zürich, Switzerland — [2]Department of Material Science and Engineering, University of Science and Technology of China, Hefei, China

Endohedral metallofullerenes (EMFs) with paramagnetic ions such as Dy^{3+} or Tb^{3+} provide a unique platform to study a spin system. Typically, magnetic interactions between neighboring carbon cages are neglected in the explanation of magnetic order above 3 K [1]. Unexpectedly, we have observed ferromagnetism at room temperature in cubic crystals of trinuclear nitrogen molecules using SQUID magnetometry. Dissolving the crystals in toluene reduces the remanence. For the dissolved $Tb_3N@C_{80}$ single molecule magnets we find hysteresis at 1.8 K without remanence, which reflects the frustrated ground state like in $Dy_3N@C_{80}$ [2]. Such a manifestation of magnetism implies a new aspect of magnetic properties of endofullerene single molecule magnets.

[1] A. Kostanyan et al. Phys. Rev. B, 101, 134429 (2020).
[2] R. Westerström et al. Phys. Rev. B, 89, 060406(R) (2014).

MA 6.6 Mon 11:00 EB 301

Hybrid Single Molecule Magnet - Metal System Studied with Nitrogen Vacancy Relaxometry — •Julian Skolaut[1], Zhewen Xu[2], Laura van Schie[2], Dominik Laible[1], Andrea Morales[3], Simon Josephy[3], Ashish Moharana[1], Eva Rentschler[1], Christian Degen[2], and Angela Wittmann[1] — [1]Johannes Gutenberg-University, Mainz, Germany — [2]ETH, Zurich, Switzerland — [3]QZabre, Zurich, Switzerland

The unceasing demand for data storage capacity continues to inspire research in the field of magnetism. Single molecule magnets (SMMs) represent a promising avenue for miniaturizing data storage units to nanometer scales.
However, integration of SMMs into devices commonly involves deposition on metal substrates, which poses many challenges. Among these, a critical concern is hybridization, which can severely alter an SMM's magnetic properties to the point of complete quenching. Here, we aim to investigate these hybridization effects on SMMs for spintronics applications.
Our investigations employ relaxometry measurements facilitated by a scanning nitrogen vacancy magnetometry setup. In this, increased magnetic noise from the SMMs leads to a reduced lifetime of the nitrogen vacancy center, providing access to the SMM's magnetic properties. We present first results of relaxometry measurements on metallacrown SMMs on gold surfaces.

15 min. break

MA 6.7 Mon 11:30 EB 301

Time-dependent density functional theory studies of a Fe(II) spin-crossover complex — •Gérald Kämmerer and Peter Kratzer — Faculty of Physics, University of Duisburg-Essen

Motivated by recent time-resolved experiments, we study the spin-state switching of a Fe(II) spin-crossover complex $Fe(pypypyr)_2$ from a diamagnetic low-spin ($S = 0$) to a paramagnetic high-spin ($S = 2$) state in the framework of density functional theory (DFT). The calculations were performed with the FHI-Aims code using PBE and HSE functionals. Due to the switching, the bond length Fe-N increases by up to 10%. In addition, excited state calculations have been performed for the electronically low spin state to understand the mechanism of light-induced switching. Molecular dynamics simulations were performed to further investigate the role of ionic motion in the switching. The financial support of the DFG within the SFB 1242 (project B02) and the computational time on the magnitUDE supercomputer system are gratefully acknowledged

MA 6.8 Mon 11:45 EB 301

Investigation of magnetic $3d$-$4f$ interaction in butterfly-shaped V_2Ln_2 complexes — •Jan Arneth[1], Xianfeng Li[2], Jonas Braun[2], Annie Powell[2], and Rüdiger Klingeler[1] — [1]Kirchhoff Institute for Physics, Heidelberg University, Germany — [2]Institute for Nanotechnology, Karlsruhe Institute for Technology, Germany

On the journey to high-performance single molecular magnets (SMM) heterometallic nanoclusters comprising $3d$ and $4f$ metal ions are of immense interest for studying the factors that govern the strength and type of intracluster magnetic coupling between the metal ions. Here we investigate the magnetic anisotropy and interactions in a family of butterfly-shaped molecular V_2Ln_2 (Ln = Y,Tb,Dy,Ho,Er,Tm,Yb) clusters where the V(III) are located on the wingtips and the Ln(III) occupy the body positions. In this series the compounds with Ln = Tb, Dy, Ho and Er show slow relaxation of the magnetisation. Combined magnetic studies and high-field/high-frequency electron paramagnetic resonance (HFEPR) spectroscopy uncover planar anisotropy of the vanadium ion and a weak antiferromagnetic V-V coupling. Furthermore, the data reveal dominant ferromagnetic V-Ln interaction which decreases with increasing number of $4f$-electrons.

MA 6.9 Mon 12:00 EB 301

Pressure-induced ordering in a highly symmetric quantum magnet DTN — •Kirill Povarov[1], David Graf[2], Andreas Hauspurg[1,3], Sergei Zherlitsyn[1], Joachim Wosnitza[1,3], Takahiro Sakurai[4], Hitoshi Ohta[5,6], Shojiro Kimura[7], Hiroyuki Nojiri[7], Ovidiu Garlea[8], Andrey Zheludev[9], Armando Paduan-Filho[10], Michael Nicklas[11], and Sergei Zvyagin[1] — [1]Hochfeld-Magnetlabor Dresden (HLD-EMFL), HZDR, Dresden — [2]National High Magnetic Field Laboratory, Tallahassee — [3]Institut für Festkörper- und Materialphysik, TU Dresden — [4]Research Facility Center for Science and Technology, Kobe U. — [5]Molecular Photoscience Research Center, Kobe U. — [6]Graduate School of Science, Kobe U. — [7]Institute for Materials Research, Tohoku U. — [8]Neutron Scattering Division, ORNL — [9]Laboratory for Solid State Physics, ETH Zürich — [10]Instituto de Fisica, U. de São Paulo — [11]Max Planck Institute for Chemical Physics of Solids

We experimentally demonstrate the pressure-induced ordering in the model tetragonal $S = 1$ quantum paramagnet $NiCl_2{\cdot}4SC(NH_2)_2$ (DTN). Employing TDO susceptibility and ultrasound techniques, we show that the spin gap vanishes and the magnetic order appears at a critical pressure $P_c = 4.2(3)$ kbar. Powder neutron diffraction reveals undistorted tetragonal symmetry at the magnetic criticality. We describe the obtained critical fields employing linear spin-wave theory and a quasi-1D numeric approximation, circumventing the quantum renormalization effects for spin-Hamiltonian parameteres. The studies are complemented by high-pressure ESR measurements.

MA 6.10 Mon 12:15 EB 301

Determination of Directions of the Magnetic Anisotropy Axes of 3d-4f Heterometallic M_2Ln_2 Single-Molecule Magnets by Inelastic Neutron Scattering — •Julius Mutschler[1], Thomas Ruppert[2], Yan Peng[2], Jacques Ollivier[3], Quentin Berrod[3], Jean-Marc Zanotti[3], Christopher E. Anson[2], Annie K. Powell[2], and Oliver Waldmann[1] — [1]Physikalisches Institut, Universität Freiburg, D-79104 Freiburg, Germany — [2]Institut of Inorganic Chemistry, Karlsruhe Institute of Technology (KIT), D-76131 Karlsruhe, Germany — [3]Institut Laue-Langevin, F-38042 Grenoble Cedex 9, France

The discovery of slow relaxation and quantum tunneling of magnetisation in single molecule magnets (SMMs) three decades ago triggered intense research into their magnetic properties. This class of molecules has been extended to heterometallic clusters containing transition metal and rare earth ions. $4f$ ions are of interest because of their large angular momentum and magnetic anisotropies but also pose a challenge in analyzing inelastic neutron scattering (INS) data. In this work, we present an INS study on non-deuterated powder samples of M_2Ln_2 butterflies with M = Fe, Al and Ln = Dy, Er, obtained with the time-of-flight disk-chopper spectrometers IN5 and IN6-SHARP at the ILL. Our analysis unveils yet another capability of powder INS: The relative directions of magnetic anisotropy axes of $4f$ ions with respect to the anisotropy axes of the $3d$ ions could be determined from the experimental data. This allows to test, e.g., ab-initio theory, in unprecedented detail.

MA 6.11 Mon 12:30 EB 301

Deep Learning based Inverse Design of Ligand-Field Parameters of Single-Molecule Magnets — •Zayan Ahsan Ali, Julius Mutschler, and Oliver Waldmann — Physikalisches Institut, Universität Freiburg, D-79104 Freiburg, Germany

Single molecule magnets (SMMs) have attracted a rich volume of research in the recent decades due to their potential applications in magnetic memory and quantum computing. Lanthanide-based SMMs in particular demonstrate promising magnetic retention due to large inherent anisotropies. Their magnetic properties can be parameterized by ligand-field theories involving a set of 27 parameters. Experimental data such as magnetization and susceptibility curves, however, are typically featureless for these materials. Multiple distinct parameter sets can describe the data to equal accuracy, making the determination of model parameters a formidable inverse problem. In this work, the over-parameterization is tackled by implementing a deep learning architecture consisting of a Variational Autoencoder (VAE) in conjunction with an Invertible Neural Network (INN). The VAE-INN architecture determines hidden system parameters of the magnetic data and subsequently relates them to multiple valid model parameters from ligand-field theory. This approach is found to offer significant advantages over conventional fitting routines, such as Levenberg-Marquardt, in terms of generalization and convergence. The study investigates and presents both the merits and the effectiveness of the VAE-INN model in producing consistent sets of ligand-field parameters for novel experimental data.

MA 6.12 Mon 12:45 EB 301

Enhancement and manipulation of quantum entanglement in three-spin clusters by non-conserving magnetization and electric field — •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 magnetoelectric 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.