

TT 13: Frustrated Magnets - Spin Liquids (joint session TT/MA)

Time: Monday 15:00–18:45

Location: Theater

TT 13.1 Mon 15:00 Theater

Designer spin liquids — ●NIC SHANNON^{1,2}, HAN YAN², OWEN BENTON³, and LUDOVIC JAUBERT⁴ — ¹TUM, Garching, Germany — ²OIST, Okinawa, Japan — ³RIKEN, Wakoshi, Japan — ⁴Universite Bordeaux, Bordeaux, France

The pyrochlore lattice has proved a rich source of spin liquids, both in theory, and in the experiment. The best known examples are “spin ices” such as Dy₂Ti₂O₇, which offer a concrete realisation of a U(1) lattice gauge theory, complete with magnetic monopole excitations. However many other spin liquid-materials are known, with many different types of phenomenology, motivating the question “what else is out there ?”

In this talk we show how a variety of different spin liquids on the pyrochlore lattice can be generated systematically, by exploiting the degeneracies which arise where different forms of order meet. As examples we present the tensor spin liquid found in models of pyrochlores with anisotropic exchange interactions [1]; the nematic spin liquid found in frustrated quantum spin ice [2,3]; and a rank-2 U(1) spin liquid found by perturbing a simple Heisenberg antiferromagnet [4]. In all cases, the predictions of the relevant gauge theory are compared with the results of Monte Carlo simulation. The relevance of these results to experiments on pyrochlore magnets, including Tb₂Ti₂O₇, is also discussed.

- [1] O. Benton *et al.*, Nat. Commun. **7**, 11572 (2016)
 [2] M. Taillefumier *et al.*, Phys. Rev. X **7**, 041057 (2017)
 [3] O. Benton *et al.*, Phys. Rev. Lett. **121**, 067201 (2018)
 [4] H. Yan *et al.*, preprint.

TT 13.2 Mon 15:15 Theater

MIEZE spectroscopy of spin dynamics and crystal field excitations in Tb₂Ti₂O₇ — ●ANDREAS WENDL¹, STEFFEN SÄUBERT^{1,2}, CHRISTIAN FRANZ², OLAF SOLTWEDEL^{1,4}, JOHANNA JOCHUM^{2,5}, PRABHAKARAN DHARMALINGAM³, ANDREW BOOTHROYD³, and CHRISTIAN PFLEIDERER¹ — ¹Technische Universität München, Garching, Germany — ²Heinz Maier-Leibnitz Zentrum (MLZ), Garching, Germany — ³Clarendon Laboratory, University of Oxford, United Kingdom — ⁴Technische Universität Darmstadt, Darmstadt, Germany — ⁵Bayerisches Geoinstitut, Bayreuth, Germany

The nature of the spin liquid ground state in the cubic rare earth pyrochlore oxide Tb₂Ti₂O₇ has been attracting great interest for many years, where recent studies suggest a prominent role of magneto-elastic crystal field - phonon interactions [1,2]. We present measurements of the spin dynamics of Tb₂Ti₂O₇ by means of the Modulation of Intensity by Zero Effort technique (MIEZE) [3], representing an implementation of high-resolution neutron spin echo suitable for depolarizing sample conditions. Our data of the intermediate scattering function cover time-scales of over seven orders of magnitude between 1 fs and 1 ns, corresponding to an energy spectrum from the meV to neV regime. We find strong paramagnetic fluctuations as well as crystal field transitions at elevated temperatures, shedding new light on the low-lying spin dynamics.

- [1] Constable *et al.*, Phys. Rev. B, **95**, 020415(R) (2017)
 [2] Fennell *et al.* Phys. Rev. Lett., **112**, 017203 (2014)
 [3] Franz and Schröder, J. Large-Scale Res. Facil. JLSRF **1**, 14 (2015)

TT 13.3 Mon 15:30 Theater

Magnetisation Avalanches in Classical Spin Ice Dy₂Ti₂O₇ — ●M. KLEINHANS¹, C. DUVINAGE¹, D. PRABHAKARAN², A. T. BOOTHROYD², and C. PFLEIDERER¹ — ¹Physik-Department, Technische Universität München, D-85748 Garching, Germany — ²Department of Physics, University of Oxford, Clarendon Laboratory, Parks Road, Oxford, OX1 3PU, United Kingdom

Spin ice attracts great interest as a state in which emergent fractionalized excitations and magnetic-field induced topological forms of order may occur [1]. We report vibrating coil magnetometry down to mK temperatures [2,3] of Dy₂Ti₂O₇, addressing the evidence of putative magnetisation avalanches in the spin-frozen state which depend sensitively in number and size on the magnetic field ramp rate, sample shape and quality. These avalanches have been interpreted in terms of magnetic monopole dynamics [4].

- [1] Castelnovo *et al.*, Nature **451**, 7174 (2008)
 [2] Krey *et al.*, PRL **108**, 257204 (2012)

- [3] Legl *et al.*, PRL **109**, 047201 (2012)
 [4] Slobinsky *et al.*, PRL **105**, 267205 (2010)

TT 13.4 Mon 15:45 Theater

Magnetization beyond the Ising limit of Ho₂Ti₂O₇ — ●L. OPPERDEN¹, T. HERRMANNSDÖRFER¹, M. UHLARZ¹, D. I. GORBUNOV¹, A. MIYATA², O. PORTUGALL², I. ISHII³, T. SUZUKI³, and J. WOSNITZA^{1,4} — ¹Hochfeld-Magnetlabor Dresden (HLD-EMFL), HZDR, Dresden, Germany — ²Laboratoire National des Champs Magnetiques Intenses (LNCMI-EMFL), Toulouse, France — ³Department of Quantum Matter, AdSM, Hiroshima University, Japan — ⁴Institut für Festkörper- und Materialphysik, TU Dresden, Germany

We report that the local Ising anisotropy in pyrochlore oxides – the crucial requirement for realizing the spin-ice state – can be broken by means of high magnetic fields. For the case of the well-established classical spin-ice compound Ho₂Ti₂O₇ the magnetization exceeds the angle-dependent saturation value of the Ising limit using ultra-high fields up to 120 T. However, even under such extreme magnetic fields full saturation cannot be achieved. Crystal-electric-field calculations can account for the measured magnetization dependence and reveal that a level crossing for two of the four ion positions leads to magnetization steps at 55 and 100 T. In addition, we show that by using a field-sweep rate in the range of the spin-relaxation time, the dynamics of the spin system can be probed. Exclusively at 25 ns/T a novel peak of the susceptibility appears around 2 T. We argue, this signals the cross-over between spin-ice and polarized correlations.

TT 13.5 Mon 16:00 Theater

The Quantum Life of Worms: Quantum Spin Ice in a [100] Magnetic Field — ●OLGA SIKORA¹, KARLO PENC², FRANK POLLMANN³, YING-JER KAO⁴, and NIC SHANNON⁵ — ¹Institute of Nuclear Physics, Polish Academy of Sciences, ul. Radzikowskiego 152, PL-31342 Kraków, Poland — ²Wigner Research Centre for Physics, H-1525 Budapest, POB 49, Hungary — ³Physics Department, Technical University Munich, 85748 Garching, Germany — ⁴Department of Physics, National Taiwan University, Taipei 10617, Taiwan — ⁵Okinawa Institute for Science and Technology Graduate University, Onna, Okinawa, 904-0495 Japan

Quantum spin ice in a magnetic field exhibits rich physics, with many open questions about possible ordered and spin-liquid states. Here we consider the case of strong [100] magnetic field, and study excitations about the maximally-polarized spin-ice state, within a model with short-range interactions. In this approach a single string of flipped spins – a “worm” – can be mapped onto an S = 1/2 XXZ chain. This mapping provides a complete understanding of a single string, exhibiting different properties in the gapped (confined) and gapless (extended) phase of the XXZ model. We further investigate the interaction between strings, using both an effective model, and large-scale variational and Green’s function Monte Carlo methods previously applied to quantum spin ice in zero field [1].

- [1] N. Shannon, O. Sikora, F. Pollmann, K. Penc and P. Fulde, Phys. Rev. Lett. **108**, 067204 (2012).

TT 13.6 Mon 16:15 Theater

Investigation of the Thermodynamic Properties of Insulating Pr-based Pyrochlores — ●J. GRONEMANN^{1,2}, T. GOTTSCHALL¹, E.L. GREEN¹, H.D. ZHOU³, A. ISLAM⁴, B. LAKE^{4,5}, and J. WOSNITZA^{1,2} — ¹Hochfeld-Magnetlabor Dresden (HLD-EMFL), HZDR, Dresden, Germany — ²Institut für Festkörper- und Materialphysik, TU Dresden, Germany — ³Univ. of Tennessee, Knoxville, USA — ⁴Helmholtz-Zentrum Berlin, Germany — ⁵Institut für Festkörperphysik, TU Berlin, Germany

Geometrically frustrated pyrochlores exhibit novel properties at low temperatures and are well-known spin-liquid candidates. In the insulating compounds Pr₂Sn₂O₇ and Pr₂Hf₂O₇ the orientation of the spins of the Pr³⁺ ions on corner-sharing tetrahedrons show dynamics beyond the spin-ice state [1]. Due to the small magnetic moment of the Pr³⁺ ion, generating only a small dipolar interaction, transverse fluctuations may have a significant influence. The spin dynamics in these materials remains unfrozen to lowest temperatures and the possibility of quantum fluctuations makes them quantum spin-liquid candidates

[2], which are expected to host a variety of emergent electrodynamic phenomena in analogy to magnetic monopoles in spin-ice. To probe the nature of the low-temperature ground state and the changes in the entropy, specific heat was measured down to 450 mK and up to 13 T.

[1] H. D. Zhou. et al., Phys. Rev. Lett. **101**, 227204 (2008)

[2] R. Sibille et al., Phys. Rev. B **94**, 024436 (2016)

TT 13.7 Mon 16:30 Theater

Giant magneto-elastic effect in d^2 pyrochlores and the formation of a spin-lattice liquid — •ANDREW SMERALD¹ and GEORGE JACKELI^{1,2} — ¹Max Planck Institut für Festkörperforschung, Heisenbergstraße 1, D-70569 Stuttgart — ²Institute for Functional Matter and Quantum Technologies, University of Stuttgart, Pfaffenwaldring 57, D-70569 Stuttgart

We discuss the idea of a giant magneto-elastic effect in frustrated magnets, and suggest that this may provide a good way to understand d^2 pyrochlore systems such as $Y_2Mo_2O_7$. We define magneto-elastic coupling as “giant” when it selects low-temperature spin configurations that are completely unexpected from the point of view of a pure spin model. This can be contrasted with the more usual case in which magneto-elastic coupling selects one or more of the otherwise extensively degenerate ground states of a frustrated magnet. In the case of $Y_2Mo_2O_7$ we propose that this mechanism results in a classical spin-lattice liquid at intermediate temperatures, in which spin and lattice degrees of freedom are intimately coupled together.

15 min. break.

TT 13.8 Mon 17:00 Theater

Intermultiplet transitions and long-range order in Sm-based pyrochlores — •VIVIANE PEÇANHA-ANTONIO¹, ERXI FENG¹, DEVASHIBHAI ADROJA², FABIO ORLANDI², XIAO SUN³, YIXI SU¹, and THOMAS BRÜCKEL³ — ¹Jülich Centre for Neutron Science (JCNS) at Heinz Maier-Leibnitz Zentrum (MLZ), Forschungszentrum Jülich GmbH, Garching, Germany — ²ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot, United Kingdom — ³Jülich Centre for Neutron Science (JCNS) and Peter Grünberg Institut (PGI), Forschungszentrum Jülich GmbH, Jülich, Germany

We present bulk and neutron scattering measurements performed on the isotopically enriched $^{154}Sm_2Ti_2O_7$ and $^{154}Sm_2Sn_2O_7$ samples. Both compounds display sharp heat capacity anomalies, at 350 mK and 440 mK, respectively. Inelastic neutron scattering measurements are employed to solve the crystalline electric field (CEF) excitations scheme, which includes transitions between the ground and first excited J multiplets of the Sm^{3+} ion. To further validate those results, the single-ion magnetic susceptibility of the compounds is calculated and compared with the experimental dc-susceptibility measured in low applied magnetic fields. It is demonstrated that the inclusion of intermultiplet transitions in the CEF analysis is fundamental to the understanding of the intermediate and, more importantly, low temperature magnetic behaviour of the Sm-based pyrochlores. Finally, the heat capacity anomaly is shown to correspond to the onset of an all-in-all-out long-range order in the stannate sample, while in the titanate a dipolar long-range order can be only indirectly inferred.

TT 13.9 Mon 17:15 Theater

Field-induced magnetic transitions in the Yb-based $J_{\text{eff}} = \frac{1}{2}$ triangular lattice antiferromagnet $NaYbO_2$ — •KIZHAKKE MALAYIL RANJITH KUMAR¹, DARYNA DMYTRIIEVA², SEUNGHYUN KIM¹, JÖRG SICHELSCHEIDT¹, HIROSHI YASUOKA¹, HANNES KÜHNE², and MICHAEL BAENITZ¹ — ¹Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany — ²Hochfeld-Magnetlabor Dresden (HLD-EMFL), HZDR, D-01314 Dresden, Germany

Spin- $\frac{1}{2}$ triangular lattice antiferromagnets (TLAF) are one of the active fields of research in condensed matter physics. The Yb^{3+} -based delafossite $NaYbO_2$ provides an ideal $J_{\text{eff}} = \frac{1}{2}$ triangular lattice motif with spin-orbit entanglement. We have synthesized the phase pure polycrystalline $NaYbO_2$ material and investigated the ground state properties. At zero field, $NaYbO_2$ exhibits no sign of magnetic long-range order down to 0.35 K, which proposes a spin liquid like ground state with strong persisting quantum fluctuations. In external magnetic fields above 2 T, it yields field-induced ordered phases. We investigated the magnetic properties in detail by magnetization, specific heat, nuclear magnetic resonance (NMR), and electron spin resonance (ESR) experiments down to 0.35 K. The results are discussed within

the extended XXZ model for bond-dependent exchange interactions on planar triangles.

TT 13.10 Mon 17:30 Theater

Spin orbit entangled planar $J = 1/2$ triangular lattice magnet $NaYbS_2$: from a putative spin liquid to field induced magnetic order — •M. BAENITZ¹, K.M. RANJITH¹, PH. SCHLENDER², J. SICHELSCHEIDT¹, B. SCHMIDT¹, H. YASUOKA¹, A.P. MACKENZIE¹, and TH. DOERT² — ¹MPI for Chemical Physics of Solids, D-01187 Dresden, Germany — ²TU Dresden, Department of Chemistry and Food Chemistry, D-01062 Dresden, Germany

Spin orbit coupling (SOC) brought significant progress to the field of quantum spin liquids (QSLs). Having strong spin orbit entanglement promote Yb-based magnets to ideal prime candidates for QSLs and as such $NaYbS_2$ is a unique model system for planar spin 1/2 triangular lattice magnetism (TLM). In contrast to $YbMgGaO_4$ [1], which shares the same space group (R-3m) and highlighted as first SOC-TLM-QSL, $NaYbS_2$ lacks inherent lattice distortions and Yb resides on a unique centrosymmetric position in the YbS_6 octahedron. Our comprehensive single crystal study combines bulk- and local- probes and identifies $NaYbS_2$ as a new spin orbit entangled TLM and putative QSL hosted on a perfect triangular lattice [2]. The application of fields along the (a,b)-plane introduces magnetic order, whereas for fields in the c-direction the system remains unaffected. We present magnetization, specific heat and NMR data down to 300 mK for both directions.

[1] J.A.M. Paddison et al., Nat. Phys. **13**, 112 (2017)

[2] M. Baenitz et al. arXiv:1809.01947 (2018)

TT 13.11 Mon 17:45 Theater

Frustrated Ising magnetism of $TmMgGaO_4$ — YUESHENG LI, •ALEXANDER A. TSIRLIN, and PHILIPP GEGENWART — EP VI, EKM, University of Augsburg, Germany

Motivated by the interesting spin-liquid physics of the triangular antiferromagnet $YbMgGaO_4$, we studied its Tm-based analog. Unlike Yb^{3+} , Tm^{3+} is a non-Kramers ion that would normally feature non-magnetic singlet as the crystal-field ground state. However, random crystal electric field (CEF) caused by the random distribution of Mg and Ga in the structure mixes two lowest-lying CEF singlets into a quasidoublet that gives rise to Ising-like pseudospins with $g_{\parallel} \simeq 13.2$ and $g_{\perp} \simeq 0$. Low-temperature thermodynamic measurements indicate three field-induced phase transitions that can be broadly understood within the $J_1^{zz} - J_2^{zz}$ Ising model on the triangular lattice, albeit with a distribution of the critical fields and underlying exchange couplings. Interestingly, only one ordered state, the $\frac{1}{3}$ -plateau below 2.5 T, shows long-range order confirmed by neutron diffraction, whereas other ordered states expected in the $J_1^{zz} - J_2^{zz}$ triangular Ising antiferromagnet seem to be only short-range in nature. Moreover, no zero-point entropy is observed.

TT 13.12 Mon 18:00 Theater

Low-energy spin excitations in the triangular-lattice quantum spin liquid candidate $YbMgGaO_4$ — •YUESHENG LI, ALEXANDER TSIRLIN, and PHILIPP GEGENWART — Experimental Physics VI, Center for Electronic Correlations and Magnetism, University of Augsburg, 86159 Augsburg, Germany

$YbMgGaO_4$ was first proposed as a perfect triangular-lattice rare-earth quantum spin liquid (QSL) candidate in 2015. After that, several scenarios, such as the spin-liquid mimicry, valence bond (VB) glass, and spin-glass, were reported in the presence of the site-mixing disorder between nonmagnetic Mg^{2+} and Ga^{3+} . Here, we critically test these scenarios by probing the low-energy spin excitations of $YbMgGaO_4$ based on the low- T magnetization and triple-axis inelastic neutron scattering (INS) experiments. Our magnetization data measured down to 40 mK speak against any conventional freezing and reinstate $YbMgGaO_4$ as a QSL candidate. The low-energy ($E \leq J_0 \sim 0.2$ meV) part of the INS continuum presents at low temperatures, but *completely* disappears upon warming the system above $T \gg J_0/k_B$. In contrast to the high-energy part at $E > J_0$ that is rooted in the breaking of nearest-neighbor VBs and persists to temperatures well above J_0/k_B , the low-energy one originates from the rearrangement of the valence bonds and thus from the propagation of unpaired spins. We further extend this picture to herbertsmithite, the QSL candidate on the kagome lattice, and argue that such a hierarchy of magnetic excitations may be a universal feature of QSLs.

TT 13.13 Mon 18:15 Theater

Randomness in the quantum spin liquid candidate κ -(BEDT-TTF)₂Cu₂(CN)₃ investigated by artificial distortion of the triangular lattice — •YOHEI SAITO¹, ANDREJ PUSTOGOW¹, ROLAND RÖSSLHUBER¹, MIRIAM ALONSO¹, MAXIM WENZEL¹, ANJA LÖHLE¹, MARTIN DRESSEL¹, TAKA AKI MINAMIDATE², NORIAKI MATSUNAGA², KAZUSHIGE NOMURA², and ATSUSHI KAWAMOTO² — ¹Physikalisches Inst., Universität Stuttgart, Germany — ²Department of Physics, Hokkaido University, Sapporo, Japan

The organic-molecular solid κ -(BEDT-TTF)₂Cu₂(CN)₃ is recognized as a quantum spin liquid candidate as it does not show magnetic ordering regardless of the large magnetic interactions. There is a debate about the importance of spin frustration on the triangular lattice and inherent randomness in the crystals. Does a suppression of geometrical frustration change the magnetic properties? To clarify that, we artificially distorted triangular lattices of κ -(BEDT-TTF)₂Cu₂(CN)₃ by donor molecular substitution that modifies exchange interactions. As a result, geometrical frustration is suppressed locally. We performed electric conductivity, dielectric spectroscopy, infrared spectroscopy, and ¹³C NMR measurements. Comparing results of non-substituted and substituted samples, we found that their magnetic fluctuation was the same as opposed to the remarkable impurity substitution effect of the conductivity. Thus, the electronic state of the (CN)₃ salt is already disordered even in the non-substituted sample, and that not only the ideal geometrical frustration but also the disorder effect should be considered.

TT 13.14 Mon 18:30 Theater

Thermal expansion studies on the spin-liquid candidate system κ -(BEDT-TTF)₂Ag₂(CN)₃ — •S. HARTMANN¹, E. GATI², Y. YOSHIDA³, G. SAITO⁴, and M. LANG¹ — ¹Physikalisches Institut, SFB/TR 49, Goethe-Uni Frankfurt, Germany — ²Ames Laboratory, Iowa State University, USA — ³Division of Chemistry, Kyoto University, Japan — ⁴Toyota Physical and Chemical Research Institute, Nagakute, Japan

The search for the realization of a quantum spin-liquid (QSL) is a major concern for condensed matter physicists since its proposal in 1973. The entangled QSL state lacks magnetic ordering down to lowest temperatures where spins continue to fluctuate even at $T = 0$ K [1]. One way to experimentally realize a QSL is magnetic frustration of geometric origin, inherent to the quasi-2D triangular lattice of the organic charge-transfer salts κ -(BEDT-TTF)₂X, known as weak Mott insulators. We present results of ultra-high-resolution thermal expansion measurements on the newly-synthesized QSL-candidate system $X = \text{Ag}_2(\text{CN})_3$. Our main finding includes pronounced broad extrema in the thermal expansion coefficient at $T \sim 18$ K along all three crystallographic directions which we assign to the effect of strong electronic correlations. The observed anomalies are qualitatively consistent with theoretical results based on the Hubbard model on a triangular lattice [2]. The directional anisotropy of the anomalies implies a ratio of the hopping integrals $t'/t < 1$.

[1] Balents, Nature 2010

[2] Kojalj, McKenzie, PRB 2015