

## TT 100: Frustrated Magnets - Pyrochlore Oxides

Time: Thursday 15:30–18:00

Location: H 3005

TT 100.1 Thu 15:30 H 3005

**Orientation Dependence of the Magnetic Phase Diagram of  $\text{Yb}_2\text{Ti}_2\text{O}_7$**  — ●STEFFEN SÄUBERT<sup>1,2</sup>, CHRISTOPHER DUVINAGE<sup>1</sup>, ALLEN SCHEIE<sup>3</sup>, JONAS KINDERVATER<sup>3</sup>, HITESH CHANGLANI<sup>3</sup>, SHU ZHANG<sup>3</sup>, SEYED KOOPPAYEH<sup>3</sup>, OLEG TCHERNYSHYOV<sup>3</sup>, COLLIN BROHOLM<sup>3,4,5</sup>, and CHRISTIAN PFLEIDERER<sup>1</sup> — <sup>1</sup>Physik Department, Technische Universität München, Germany — <sup>2</sup>Heinz Maier-Leibnitz Zentrum, Technische Universität München, Germany — <sup>3</sup>Institute for Quantum Matter and Department of Physics and Astronomy, Johns Hopkins University, USA — <sup>4</sup>NIST Center for Neutron Research, National Institute of Standards and Technology, USA — <sup>5</sup>Department of Materials Science and Engineering, Johns Hopkins University, USA

The magnetic pyrochlore oxide  $\text{Yb}_2\text{Ti}_2\text{O}_7$  received a lot of attention in recent years, as strong transverse quantum fluctuations significantly influence the system, and since the nature of its ground state is still under debate to host a spin liquid quantum state, i.e. quantum spin ice, at low temperatures. We report the orientation dependence of the magnetic phase diagram of  $\text{Yb}_2\text{Ti}_2\text{O}_7$ , inferred from magnetometry down to millikelvin temperatures, and further address the question of the ground state of  $\text{Yb}_2\text{Ti}_2\text{O}_7$ . The magnetic phase diagram for externally applied field shows an unusual field dependence of a first-order phase boundary, notably an applied field initially increases the ordering temperature when applied parallel to the crystalline  $\langle 111 \rangle$  or  $\langle 110 \rangle$  axes. This unusual field dependence is absent for field along  $\langle 100 \rangle$ . The zero-field ground state was found to be of ferrimagnetic order, with spins slightly splayed away from one of the six  $\langle 100 \rangle$  directions.

TT 100.2 Thu 15:45 H 3005

**The Magnetic Excitations in the Ground State of  $\text{Yb}_2\text{Ti}_2\text{O}_7$**  — ●VIVIANE PEÇANHA-ANTONIO<sup>1,2</sup>, ERXI FENG<sup>1</sup>, YIXI SU<sup>1</sup>, FRANZ DEMMEL<sup>3</sup>, and THOMAS BRÜCKEL<sup>1,4</sup> — <sup>1</sup>Jülich Centre for Neutron Science (JCNS) at Heinz Maier-Leibnitz Zentrum (MLZ), Forschungszentrum Jülich GmbH, Lichtenbergstr. 1, D-85747 Garching, Germany — <sup>2</sup>Physik-Department, Technische Universität München, D-85747 Garching, Germany — <sup>3</sup>ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot OX11 0QX, United Kingdom — <sup>4</sup>Jülich Centre for Neutron Science (JCNS) and Peter Grünberg Institut (PGI), Forschungszentrum Jülich GmbH, D-52425 Jülich, Germany

The nature of the zero-field ground state of  $\text{Yb}_2\text{Ti}_2\text{O}_7$  remains an enigma within the pyrochlore titanate series. The disparate results are attributed to subtle changes in the sample stoichiometry, which seems to tune the magnetic order of different samples across a phase boundary. We report a study on the zero-field ground state of a powder sample of this pyrochlore. A sharp heat capacity anomaly at  $T_c = 280$  mK is accompanied by a quasicollinear ferromagnetic order with a magnetic moment of  $0.87(2)\mu_B$ . Our high-resolution inelastic neutron scattering measurements show that, upon cooling, an inelastic continuum of excitations at  $\sim 0.6$  is observed to persist from at least 2.5 K down to the lowest reached temperatures. Below  $T_c$ , the coexistence of sharp gapped low-energy magnetic excitations with a remnant quasielastic contribution evidences that spin fluctuations persist despite the long-range magnetic order.

TT 100.3 Thu 16:00 H 3005

**Spin dynamics in the spin ice  $\text{Ho}_2\text{Ti}_2\text{O}_7$  as measured by MIEZE spectroscopy** — ●ANDREAS WENDL<sup>1</sup>, S. SÄUBERT<sup>1,2</sup>, C. FRANZ<sup>2</sup>, P. DHARMALINGAM<sup>3</sup>, A. BOOTHROYD<sup>3</sup>, and C. PFLEIDERER<sup>1</sup> — <sup>1</sup>Technische Universität München, Garching, Germany — <sup>2</sup>Heinz Maier-Leibnitz Zentrum (MLZ), Garching, Germany — <sup>3</sup>Clarendon Laboratory, University of Oxford, United Kingdom

In the cubic rare-earth pyrochlore  $\text{Ho}_2\text{Ti}_2\text{O}_7$  the combination of a tetrahedral Ho-atom sub-lattice with strong local Ising anisotropies leads to a spin arrangement satisfying the spin ice rules at low temperatures [1]. At high temperatures the combination of crystal fields and phononic states creates rich physics: Thermally driven transitions of the ground state doublet deviate from an Arrhenius law [2] as phonon mediated spin-flipping becomes relevant[3]. Furthermore, crystal field transitions between excited states appear once these states are populated at sufficient temperatures [3,4]. We report a study of the high temperature spin flip excitations in  $\text{Ho}_2\text{Ti}_2\text{O}_7$  employing the high-resolution neutron spin echo technique MIEZE. Our measurements provide unprecedented information on the spin dynamics measured

over five magnitudes in time, thus allowing to close the gap between TAS [2,3] and NSE [2], and extending it towards lower momentum transfers. Our results agree with refs [2-4] and further confirm the single ion character of spin fluctuations.

- [1] Gardner, Rev. Mod. Phys., **82**, 53 (2010)
- [2] Ehlers, J Phys: Condens. Matter, **16**, S635 (2003)
- [3] Ruminy, Phys. Rev. B, **95**(6), 60414 (2017)
- [4] Ruminy, Phys. Rev. B, **94**(2), 24430 (2016)

TT 100.4 Thu 16:15 H 3005

**Inverted hysteresis within the antiferromagnetic all-in-all-out state of the pyrochlore  $\text{Nd}_2\text{Hf}_2\text{O}_7$**  — ●L. OPPERDEN<sup>1,2</sup>, T. BILITEWSKI<sup>3</sup>, J. HORNING<sup>1,2</sup>, T. HERRMANSDÖRFER<sup>1</sup>, A. SAMARTZIS<sup>4,5</sup>, A. T. M. N. ISLAM<sup>4</sup>, V. K. ANAND<sup>4</sup>, B. LAKE<sup>4,5</sup>, R. MOESSNER<sup>3</sup>, and J. WOSNITZA<sup>1,2</sup> — <sup>1</sup>Hochfeld-Magnetlabor Dresden (HLD-EMFL), HZDR, Dresden, Germany — <sup>2</sup>Institut für Festkörper- und Materialphysik, TU Dresden, Germany — <sup>3</sup>Max-Planck-Institut für Physik komplexer Systeme, Dresden, Germany — <sup>4</sup>Abteilung Quantenphänomene in neuen Materialien, HZB, Berlin, Germany — <sup>5</sup>Institut für Festkörperphysik, TU Berlin, Germany

We report the observation of an anisotropic and inverted hysteresis loop in the antiferromagnetic all-in-all-out ordered phase of  $\text{Nd}_2\text{Hf}_2\text{O}_7$  having a negative remnant magnetization. The hysteresis emerges once exceeding a characteristic magnetic-field strength  $H^*(T)$  below the Néel temperature. The very unusual appearance of a negative remnant magnetization is observed for a field parallel to the  $[111]$  and  $[110]$  direction. However, for field parallel to  $[001]$  no hysteresis can be seen. For this orientation the projection of the field onto all four local spin directions is equal and, hence, both realizations of the all-in-all-out state gaining equal Zeeman energy through a canting of their spins. We show further, that the underlying all-in-all-out phase is established in  $\text{Nd}_2\text{Hf}_2\text{O}_7$  for temperatures below  $T_N = 0.48$  K and persists up to fields of 0.27 T. We account for the inverted hysteresis in terms of a theory of uncompensated domain-wall spins of spherical domains forming inside a fully polarized single-domain state.

TT 100.5 Thu 16:30 H 3005

**Spin dynamics of the ordered dipolar-octupolar pseudospin pyrochlore  $\text{Nd}_2\text{Hf}_2\text{O}_7$**  — ●ALEXANDROS SAMARTZIS<sup>1,2</sup>, JIANHUI XU<sup>1</sup>, VIVEK K. ANAND<sup>1</sup>, NAZMUL A.T.M. ISLAM<sup>1</sup>, JACQUES OLLIVIER<sup>3</sup>, and BELLA BELLA<sup>1,2</sup> — <sup>1</sup>Helmholtz-Zentrum Berlin — <sup>2</sup>Technical University Berlin — <sup>3</sup>Institut Laue-Langevin, Grenoble, France

The rare earth pyrochlore magnets have been extensively studied during the last decades due to their quintessential lattice for frustration which leads to exotic ground states with strong anisotropy. From this lattice the competition of crystal field, super-exchange and dipolar interactions can result in novel states, such as spin liquid, spin ice etc. The  $\text{Nd}^{3+}$  ion, on such a lattice has a Kramers doublet ground state with a 'dipolar-octupolar' wavefunction leading to a fascinating phase diagram. Here we report first results of inelastic scattering on a new Nd-based pyrochlore,  $\text{Nd}_2\text{Hf}_2\text{O}_7$ . Recent macroscopic measurements have revealed an ordered all-in - all-out AFM ground state with slow spin dynamics and a strongly reduced magnetic moment due to local fluctuations. Motivated by these interesting results, we performed low energy Inelastic Neutron scattering on a large single crystal grown by the floating zone technique. The results reveal a long range magnetic order below  $T=600$ mK. The excitations form a gapped flat band at energy DE 0.1meV reflecting the pinch-point pattern of a Coulombic phase. Above the flat band, collective dispersive excitations emerge from the pinch points. Linear spin-wave theory was used to determine the exchange parameters (Ja) applied in an appropriate Hamiltonian.

15 min. break.

TT 100.6 Thu 17:00 H 3005

**Spin freezing in disordered pyrochlore magnets probed by NMR** — RAJIB SARKAR<sup>1</sup>, ●FELIX BRÜCKNER<sup>1</sup>, JASON W. KRIZAN<sup>2</sup>, ROBERT J. CAVA<sup>2</sup>, and HANS-HENNING KLAUSS<sup>1</sup> — <sup>1</sup>Institut für Festkörper- und Materialphysik, Technische Universität Dresden, Germany — <sup>2</sup>Department of Chemistry, Princeton University, USA

The frustrated pyrochlore magnets  $\text{NaACo}_2\text{F}_7$  ( $A = \text{Ca}^{2+}, \text{Sr}^{2+}$ )

exhibit a mixed A-site with a random distribution of  $\text{Na}^+$  and  $\text{Ca}^{2+}/\text{Sr}^{2+}$ . To investigate the effect of the resulting bond-disorder, we performed  $^{23}\text{Na}$  and  $^{19}\text{F}$  NMR experiments. While the Curie-Weiss temperature is  $\sim 140$  K ( $A = \text{Ca}$ ) respective  $\sim 130$  K ( $\text{Sr}$ ), the spin freezes at around 3 K, which gives high frustration indices of around 45. In fact, the  $^{23}\text{Na}$  and  $^{19}\text{F}$  broaden substantially below 3.6 K accompanied by a considerable reduction of the NMR signal intensity. A progressive slow-down of spin fluctuations is observed as per a BPP-like curvature of the  $^{19}\text{F}$  spin-lattice relaxation rate. Eventually this ends up in a spin frozen state below 3.6 K in  $\text{NaCaCo}_2\text{F}_7$ . The hyperfine coupling to the magnetic moments increases significantly in this region. In addition to that, we present a simulation of  $^{23}\text{Na}$  NMR spectra and compare it to results of a DFT calculation.

[1] Phys. Rev. B 89, 214401 (2014)

[2] Phys. Rev. B 95, 144414 (2017)

[3] R. Sarkar et al. (accepted in PRB)

TT 100.7 Thu 17:15 H 3005

**Features of quantum spin ice in pyrochlore  $\text{Nd}_2\text{Zr}_2\text{O}_7$**  — •JIANHUI XU<sup>1,2</sup>, A. T. M. NAZMUL ISLAM<sup>1</sup>, OWEN BENTON<sup>3</sup>, GEORG EHLERS<sup>4</sup>, and BELLA LAKE<sup>1,2</sup> — <sup>1</sup>Helmholtz-Zentrum Berlin für Materialien und Energie, Hahn-Meitner-Platz 1, 14109 Berlin, Germany — <sup>2</sup>Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstraße 36, D-10623 Berlin, Germany — <sup>3</sup>RIKEN Center for Emergent Matter Science (CEMS), Wako, Saitama 351-0198, Japan — <sup>4</sup>Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA

Magnetic pyrochlore oxides are intensively studied in the field of frustrated magnetism and the pyrochlores with Ising-anisotropic light rare earth are candidates for quantum spin ice. The recent studies on  $\text{Nd}_2\text{Zr}_2\text{O}_7$  show that the single-ion ground state of  $\text{Nd}^{3+}$  is a well-isolated Kramers doublet with Ising anisotropy and the collective ground state is an antiferromagnetic order. Inelastic neutron scattering shows magnetic excitations containing a flat gapped ice-like mode and dispersive branches. The analyses based on spin wave theory indicate quantum moment fragmentation and yield a pseudospin-1/2 Hamiltonian which suggests quantum spin ice state above the ordering temperature (0.4K). Here we show that the extracted spin Hamiltonian gives a qualitative description of the macroscopic properties of  $\text{Nd}_2\text{Zr}_2\text{O}_7$  based on mean-field and Monte Carlo simulations. We also present single-crystal inelastic neutron scattering data above  $T_N$  at 450mK and compare with the calculated spinon scattering.

TT 100.8 Thu 17:30 H 3005

**Frozen state and persistent spin dynamics of new kagome compound  $\text{Fe}_4\text{Si}_2\text{Sn}_7\text{O}_{16}$ : A  $\mu\text{SR}$  and AC-susceptibility study** — •S. DENGRE<sup>1</sup>, R. SARKAR<sup>1</sup>, J.-C. ORIAN<sup>2</sup>, C. BAINES<sup>2</sup>, L. OPPERDEN<sup>3</sup>, M. UHLARZ<sup>3</sup>, T. HERRMANNSDÖRFER<sup>3</sup>, T. SÖHNEL<sup>4</sup>,

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We present the results of a new kagome compound  $\text{Fe}_4\text{Si}_2\text{Sn}_7\text{O}_{16}$  as probed by bulk AC-susceptibility and  $\mu\text{SR}$  experiments. Zero field (ZF)  $\mu\text{SR}$  spectra shows the presence of two relaxation channels with faster and slower relaxation rates ( $\lambda_1, \lambda_2$ ) respectively. A peak in both  $\lambda_1, \lambda_2$  is observed in the temperature range of 2-3 K associated with the static magnetism.  $\lambda_1$  exhibits a constant value below 1.5 K down to 270 mK indicating the presence of dynamic magnetism in the system. In AC-susceptibility, we observe a frequency dependent broad maximum which shifts from 3.5 K to 6 K (1.1 KHz). At low temperature below 0.15 K an upturn in the AC-susceptibility is evidenced. The combination of AC-susceptibility and  $\mu\text{SR}$  data suggests the presence of two relevant energy scales in  $\text{Fe}_4\text{Si}_2\text{Sn}_7\text{O}_{16}$ .

TT 100.9 Thu 17:45 H 3005

**Magnetic semimetallic state in pyrochlore ruthenate  $\text{Cd}_2\text{Ru}_2\text{O}_7$**  — •MARIAN BLANKENHORN<sup>1</sup>, TOMOHIRO TAKAYAMA<sup>2</sup>, JÜRGEN NUSS<sup>2</sup>, ROBERT DINNEBIER<sup>2</sup>, ALEXANDER YARESKO<sup>2</sup>, and HIDENORI TAKAGI<sup>1,2</sup> — <sup>1</sup>University of Stuttgart, Stuttgart, Germany — <sup>2</sup>Max Planck Institute for Solid State Research, Stuttgart, Germany

In pyrochlore oxides with the chemical composition  $A_2B_2O_7$  strong geometrical frustration gives rise to interesting phenomena such as spin-ice behavior. Metallic pyrochlore oxides also show a variety of electronic phases including superconductivity in  $\text{Cd}_2\text{Re}_2\text{O}_7$  and a metal-insulator transition (MIT) in  $\text{Tl}_2\text{Ru}_2\text{O}_7$ . While many  $\text{Ru}^{4+}$  pyrochlores have been intensively studied, only three  $\text{Ru}^{5+}$  pyrochlores,  $A_2\text{Ru}_2\text{O}_7$  ( $A = \text{Hg}, \text{Ca}, \text{Cd}$ ), are known so far. While  $\text{Ca}_2\text{Ru}_2\text{O}_7$  remains metallic at low temperatures showing spin-glass behavior,  $\text{Hg}_2\text{Ru}_2\text{O}_7$  undergoes a MIT at around 100 K. For  $\text{Cd}_2\text{Ru}_2\text{O}_7$  the formation of a SDW was proposed based on a drop of magnetic susceptibility and an anomaly in resistivity but no detailed information is available. We obtained single crystals of  $\text{Cd}_2\text{Ru}_2\text{O}_7$  by high pressure synthesis. Both magnetic susceptibility and resistivity exhibit a sharp drop at around 105 K. This transition accompanies a loss of carriers, indicating a low-carrier metallic state at low temperatures. The LDA + U band calculation suggests an antiferromagnetic semimetallic ground state with possibly all-in all-out magnetic order. We argue that  $\text{Cd}_2\text{Ru}_2\text{O}_7$  displays a unique transition where magnetic order induces a coherent semimetallic state out of an incoherent paramagnetic bad-metal.