TT 3: Correlated Electrons: Frustrated Magnets - Pyrochlore Systems and Iridates

Time: Monday 9:30–13:00 Location: H20

TT 3.1 Mon 9:30 H20

Persistent spin dynamics in $NaCaCo_2F_7$ as evidenced by μSR — •Sascha Albert Bräuninger[1], Rajib Sarkar[1], Jason W. Krizan[2], Shanu Dengre[1], Philipp Materne[1], Christopher Baines[3], Hubertus Luetkens[3], Robert J. Cava[2], and Hans-Henning Klauss[1] — [1]Institute for Solid State Physics, TU Dresden, D-01069, Germany — [2]Department of Chemistry, Princeton University, Princeton, NJ 08544, USA — [3]Laboratory for Muon-Spin Spectroscopy, Paul Scherrer Institute, CH-5232 Villigen, Switzerland

The fluoride pyrochlore $NaCaCo_2F_7$ is a newly discovered frustrated pyrochlore with a frustration index of $f = \frac{|\theta_{CW}|}{T_f} \approx 56$. While recent NMR experiments on $NaCaCo_2F_7$ suggested a spin frozen state below 3 K, neutron scattering experiments on the other hand proposed XY like antiferromagnetic spin clusters at low energies. We present μSR studies on $NaCaCo_2F_7$. Present results indicate the slowing down of the magnetic (spin) fluctuations upon cooling towards the NMR and neutron scattering spin frozen state temperature of $T_{sf} \approx 3.0$ K. The μSR relaxation rate increases slightly below this frozen state, and remains constant down to 20 mK. In the μSR window there is no indication of static magnetism in $NaCaCo_2F_7$. In longitudinal field (100-4000 G) the relaxation rate do not vary indicating that the spin fluctuations are dynamic, and this is persistent even at $T \approx 20$ mK. While persistent spin dynamics (PSD) appears to be a generic feature of frustrated magnetic systems, it is not clear so far for the present case whether this is associated with quantum fluctuations, spin-liquid physics, or some other effect.

TT 3.2 Mon 9:45 H20

Unusual spin frozen state in a frustrated pyrochlore system $NaCaCo_2F_7$ as observed by NMR — •R. Sarkar[1], J. W. Krizan[2], F. Brückner[1], R. J. Cava[2], and H.-H Klauss[1] — [1]IFP, TU Dresden, D-01069 Dresden, Germany — [2]Department of Chemistry, Princeton University, Princeton, NJ 08544, USA

We present ^{23}Na -and ^{19}F NMR results on the magnetically frustrated pyrochlore $NaCaCo_2F_7$ with a frustration index of $f = \theta_{CW}/T_f \sim 56$. Recent neutron scattering experiments proposed XY like antiferromagnetic spin clusters at low energies in $NaCaCo_2F_7$. ^{23}Na NMR -spectra reveal the presence of two magnetically non equivalent Na sites in conjunction with the local Co^{2+} spin structure. Below 3.6 K both the ^{23}Na -and ^{19}F spectra broaden due to the formation of static spin correlations. A huge reduction of the ^{19}F -and ^{23}Na NMR signal intensity hints at a quasi-static field distribution in $NaCaCo_2F_7$ in this regime. The ^{19}F spin-lattice relaxation rate $^{19}(1/T_1)$ exhibits a peak at around 2.9 K, at the same temperature range where ac and dc susceptibility data show a broad maximum. The character of the spin fluctuation appears to be isotropic. The overall temperature dependence of $^{19}(1/T_1)$ can be described by the BPP theory considering a fluctuating hyperfine field with an autocorrelation function. The correlation time of the autocorrelation function exhibits an activation behavior further indicating the spin-frozen state. While the present NMR studies suggest the spin frozen state at low temperatures, μSR investigations however reveal the presence of so called persistent spin dynamics down to 20 mK implying an exotic ground state in $NaCaCo_2F_7$.

TT 3.3 Mon 10:00 H20

Magnetoelastic properties of the quantum-spin-ice candidate $Yb_2Ti_2O_7$ — •T. Stöter[1,2,3], M. Doerr[1,2], S. Granovsky[1,2], Z.S. Wang[1,2,3], S. Erfanifam[3], E. Green[3], S. Zherlitsyn[1,3], J. Wosnitza[1,2,3], A. Maljuk[1,4], and S. Wurmehl[1,4] — [1]SFB 1143 — [2]TUD/IFP, Dresden — [3]HZDR, Dresden — [4]IFW, Dresden

Intriguing phenomena such as the occurrence of magnetic monopoles and a wide variety of ground states are associated to magnetic frustration. In a number of cases, elastic effects, e.g. lattice distortions, may result in the lifting of degeneracies or the appearance of new magnetic states. The rare-earth titanate $Yb_2Ti_2O_7$, where the magnetic Yb^{3+} ions form a pyrochlore spin network, is a prime example of a geometrically frustrated material, with numerous field-induced phases and strong ferromagnetic correlations below 170 mK. In order to characterize the magneto-elastic coupling in this material, we have investigated the thermal expansion, magnetostriction, and sound propagation in different dilution refrigerators between 60 mK and 1.5 K and large applied magnetic fields. At around 170 mK we find distinct anomalies in the expansion coefficient, acoustic properties, as well as the specific heat. Lattice anomalies in field hint to additional low temperature phases.

TT 3.4 Mon 10:15 H20

Suppression of Pauling's residual entropy in dilute spin ice $(Dy_{1-x}Y_x)_2Ti_2O_7$ — •S. Scharffe[1], O. Breunig[1], V. Cho[1], P. Laschitzky[1], M. Valldor[1,2], J. F. Welter[1], and T. Lorenz[1] — [1]II. Physikalisches Institut, Universität zu Köln, Germany — [2]Max-Planck-Institut für Chemische Physik fester Stoffe, Dresden, Germany

The spin ice $Dy_2Ti_2O_7$ is a geometrically frustrated spin system consisting of corner-sharing tetrahedra with an Ising anisotropy that aligns the spins along their local easy axes in the {111} direction. In the ground state configuration two spins point into and two out of each tetrahedron. The entropy of $Dy_2Ti_2O_7$ reveals a plateaulike feature close to Pauling's residual entropy around 0.5 K derived originally for water ice, but a distinct expansion towards lower temperature is prevented by ultraslow thermal equilibration. We present specific-heat data of $(Dy_{1-x}Y_x)_2Ti_2O_7$ and analyze the influence of nonmagnetic yttrium dilution on the low-temperature entropy. We find that these ultraslow thermal equilibration rapidly vanishes with increasing x, the low-temperature entropy systematically decreases, and its temperature dependence strongly increases[1]. From our data, a nondegenerate ground state can be derived for $(Dy_{1-x}Y_x)_2Ti_2O_7$ with intermediate dilution. This is in contrast to an expected zero-temperature residual entropy obtained from a generalization of Pauling's theory for dilute spin ice, but is supported by Monte Carlo simulations which are also compared to our results.

This work was supported by the DFG via project LO 818/2-1.

[1] Scharffe et al., PRB, **92**, 180405(R) (2015)

TT 3.5 Mon 10:30 H20

Determination of the spin Hamiltonian in the pyrochlore $Lu_2V_2O_7$ — •Kira Riedl[1], Harald O. Jeschke[1], Michel J.P. Gingras[2,3,4], and Roser Valenti[1] — [1]Institut für Theoretische Physik, Goethe-Universität Frankfurt, Max-von-Laue-Str. 1, 60438 Frankfurt am Main, Germany — [2]Department of Physics and Astronomy, University of Waterloo, Ontario, N2L 3G1, Canada — [3]Perimeter Institute for Theoretical Physics, Waterloo, Ontario, N2L 2Y5, Canada — [4]Canadian Institute for Advanced Research, 180 Dundas Street West, Suite 1400, Toronto, ON, M5G 1Z8, Canada

In the pyrochlore $Lu_2V_2O_7$ the vanadium ions form corner-sharing spin 1/2 tetrahedra. In order to find the corresponding spin Hamiltonian which captures the essential physics of the investigated compound we performed a tight-binding fit on the vanadium d orbitals using density functional theory. Since there is evidence that the Dzyaloshinskii-Moriya interaction (DMI) is important in this system, we considered spin-orbit coupling effects within our calculations.

A fitting procedure to the relativistic band structure enabled us to determine the strength of the spin-orbit coupling. In a second step, we calculated the energy parameters in the spin Hamitonian with the method of exact diagonalization and projection on low energy states. We were therefore able to evaluate the Heisenberg exchange, the DMI, and the symmetric tensor, only using *ab initio* information and reasonable values for the Hubbard interaction as well as for the Hund's coupling. Comparison with recent experimental results will be discussed.

15 min. break

TT 3.6 Mon 11:00 H20

Neutron scattering investigation of rare earth pyrochlore iridates and hafnates — •Erxi Feng[1], Yixi Su[1], Thomas Wolf[2], and Thomas Brueckel[3,1] — [1]Jülich Centre for Neutron Science JCNS, Forschungszentrum Jülich GmbH, Outstation at MLZ, D-85747 Garching, Germany — [2]Institut für Festkörperphysik, Karlsruhe Institute of Technology KIT, D-76021 Karlsruhe, Germany — [3]Jülich Centre for Neutron Science JCNS and Peter Grünberg Institut PGI, JARA- FIT, Forschungszentrum Jülich GmbH, D-52425 Jülich, Germany

Pyrochlore iridates and hafnates $A_2B_2O_7$ (A= rare earth ion, B=Ir, Hf), in which both the A-site and the B-site ions form corner-sharing

tetrahedra, have recently attracted considerable research interests due to the presence of both strong spin-orbit coupling and geometrical frustration. $A_2B_2O_7$ displays an intriguing metal-to-insulator transition (MIT) driven by both thermodynamics and chemical pressure, which is suggested to coincide with possible magnetic order at the Ir^{4+} site. Moreover, the two sublattices of A^{3+} and Ir^{4+} might be magnetically coupled thus leading to novel magnetic behaviors. As a counterpart, $A_2B_2O_7$ compounds are insulator and the Hf4+ ion is nonmagnetic. Pyrochlore iridates and hafnates powders with light rare earth Pr and Nd were synthesized by standard solid state reaction. Combined X-ray and neutron powder diffraction refinements suggest that the samples are stoichiometric. In this presentation, our recent investigations of the magnetic ground states of these compounds via comprehensive neutron scattering techniques will be reported.

TT 3.7 Mon 11:15 H20

First-principles study of strong correlation effects in pyrochlore iridates — •Hiroshi Shinaoka[1], Shintaro Hoshino[2], Matthias Troyer[3], and Philipp Werner[4] — [1]Department of Physics, Saitama University, Japan — [2]Department of Basic Science, The University of Tokyo, Japan — [3]Theoretische Physik, ETH Zuerich, Switzerland — [4]Department of Physics, University of Fribourg, Switzerland

The pyrochlore iridates $A_2Ir_2O_7$ (A=Pr, Nd, Y, etc.) are an ideal system to study fascinating phenomena induced by strong electron correlations and spin-orbit coupling. In this talk, we study strong correlation effects in the prototype compound $Y_2Ir_2O_7$ using the local density approximation and dynamical mean-field theory (LDA+DMFT) [1]. We map out the phase diagram in the space of temperature, on-site Coulomb repulsion U, and filling. Consistent with experiments, we find that an all-in/all-out ordered insulating phase is stable for realistic values of U. We reveal the importance of the hybridization between $j_{eff} = 1/2$ and $j_{eff} = 3/2$ states under the Coulomb interaction and trigonal crystal field. We demonstrate a substantial band narrowing in the paramagnetic metallic phase and non-Fermi liquid behavior in the electron/hole doped system originating from long-lived quasi-spin moments induced by nearly flat bands. We further compare our results with recent experimental results of $Eu_2Ir_2O_7$ under hydrostatic pressure [2].

[1] H. Shinaoka, S. Hoshino, M. Troyer, P. Werner, PRL **115**, 156401 (2015)
[2] G. Prando, R. Dally, W. Schottenhamel, Z. Guguchia, S.-H. Baek, R. Aeschlimann, A. U. B. Wolter, S. D. Wilson, B. Büchner, M. J. Graf, arXiv:1511.03037

TT 3.8 Mon 11:30 H20

Metal-insulator transition of pyrochlore iridates and their topological properties — •Hongbin Zhang[1,2], Kristjan Haule[2], and David Vanderbilt[2] — [1]Materialwissenschaft, TU Darmstadt Alarich-Weiss-Straße 2, 64287 Darmstadt, Germany — [2]Department of Physics and Astronomy, Rutgers University, 136 Frelinghuysen Road, NJ-08854, USA

The interplay of spin-orbit coupling and electronic correlations can lead to many fascinating physical properties, where iridates are a promising playground. Combining density functional theory (DFT) and embedded dynamical mean-field theory (DMFT) methods, we study the metal-insulator transition in $R_2Ir_2O_7$ (R=Y, Eu, Sm, Nd, Pr, and Bi) and the topological nature of the insulating compounds. Accurate free energies evaluated using the charge self-consistent DFT+DMFT method reveal that the metal-insulator transition occurs for an A-cation radius between that of Nd and Pr, in agreement with experiments. The all-in-all-out magnetic phase, which is stable in the Nd compound but not the Pr one, gives rise to a small Ir^{4+} magnetic moment of $\approx 0.5\mu_B$ and opens a sizable correlated gap. We demonstrate that within this state-of-the-art theoretical method, the insulating bulk pyrochlore iridates are topologically trivial.

TT 3.9 Mon 11:45 H20

Specific heat study of the iridium double perovskite Sr_2YIrO_6 — •Laura T. Corredor, Kaustuv Manna, Gizem Aslan Cansever, Sebastian Gass, Andreas Zimmermann, Tushar Dey, Christian Blum, Andrey Maljuk, Sabine Wurmehl, Anja Wolter, and Bernd Büchner — Leibniz Institute for Solid State and Materials Research IFW, Institute for Solid State Research, 01069 Dresden, Germany

Recently, Mott insulators with a d^4 electronic configuration were predicted to show superexchange-driven quantum phase transitions. Double perovskites $R_2MM'O_6$ with M'^{3+} ion, yielding a formal oxidation state of Ir^{5+} with $5d^4$ electronic configuration, may be candidates to verify or discard such transitions and its impact on the magnetic structure. According to the strong spin-orbit coupling J_{eff} model, a non-magnetic ground state is expected. Such material is realized in Sr_2YIrO_6. Nevertheless, it is claimed [1] that a strong non-cubic crystal field together with a "intermediate-strength" spin-orbit coupling, would lead to a different ground state configuration and to antiferromagnetic behavior with $T_N = 1.3$ K. Also, anomalies in the specific heat were associated to this novel magnetism. In this work, we present magnetic and thermodynamic characterization of Sr_2YIrO_6 single crystals. No long magnetic order was found. The magnetic contribution to the specific heat was calculated, finding a Schottky anomaly due to magnetic impurities. Further analysis suggests non-negligible spin correlations, which nonetheless, are not associated with long range magnetic ordering.

[1] G.Cao et al., PRL **112**, 056402 (2014).

TT 3.10 Mon 12:00 H20

On the search for magnetic correlations in double perovskites — •Franziska Hammerath[1,2], Rajib Sarkar[1], Sirko Kamusella[1], C. Baines[3], H.-H. Klauss[1], T. Dey[2], Gizem Aslan Cansever[2], Kaustuv Manna[2], Andreas Zimmermann[2], Andrey Maljuk[2], Mihai Sturza[2], Dmitriy Efremov[2], Sabine Wurmehl[2], and Bernd Büchner[2] — [1]Institute for Solid State Physics, Dresden Technical University, TU Dresden, 01062 Dresden, Germany — [2]IFW Dresden, Institute for Solid State Research, PF 270116, 01171 Dresden, Germany — [3]Laboratory for Muon Spin Spectroscopy, Paul Scherrer Institute, CH-5232 Villigen, PSI, Switzerland

The cubic double perovskite Ba_2YIrO_6 has been investigated by the local probe techniques NMR and μSR. Both methods confirm the absence of long range order in this compound, but observe signatures of magnetic correlations: The NMR spin-lattice relaxation rate suggests the presence of growing magnetic correlations at low temperatures. An increase of the μSR spin-lattice relaxation rate confirms the presence of weak magnetism. These findings cannot be explained by the recently suggested excitonic type of magnetism [1], but also go beyond a simple nonmagnetic ground state picture of the $5d^4$ ($J_{eff} = 0$) electronic configuration of Ir^{5+}. In the monoclinic analog Sr_2YIrO_6, the NMR line width and spin-lattice relaxation rates reveal a nonmagnetic behavior, in contrast to a first report [2], but in line with a recent study [3].

[1] G. Khaliullin, PRL **111**, 197201 (2013).
[2] G. Cao *et al.*, PRL **112**, 056402 (2014).
[3] B. Ranjbar *et al.*, Inorg. Chem. **54**, 10468 (2015).

TT 3.11 Mon 12:15 H20

High-field multi-frequency ESR spectroscopy of La_2CuIrO_6 — •Stephan Fuchs[1], Vladislav Kataev[1], Kaustuv Manna[1], Sabine Wurmehl[1], Anup Kumar Bera[3], Andrey Malyuk[1], and Bernd Büchner[1,2] — [1]Leibniz-Institut für Festkörper- und Werkstoffforschung (IFW) Dresden,D-01171 — [2]Institut für Festkörperphysik, Technische Universität Dresden,D-01062 — [3]Helmholtz-Zentrum Berlin für Materialien und Energie, 14109 Berlin, Germany

We will present the electron spin resonance results of the double perovskite La_2CuIrO_6. This material provides a playground to examine the magnetic interactions in a 5d transition metal oxide with strong spin-orbit coupling. Measurements of the static magnetization M(T,H) show an antiferromagnetic ordering at $T_{AFM} = 74$ K and a weak ferromagnetic moment below 54 K. ESR measurements of the powder sample were carried out for several temperatures and frequencies to determine the g-factor and the magnetic exaction gap. Our goal is to identify the origin of the ferromagnetic contribution with ESR. We observe an opening of the ferromagnetic gap at T=93 K ($> T_{AFM}$) which continuously develops over the T_{AFM} down to low temperature. The complex interaction of the Cu- and Ir-spin gives rise to the continuous shift of the g-factor: By decreasing the temperature, the Ir spins are getting progressively more involved in the resonance of the statically ordered Cu spin lattice due to exchange coupling between the two sublattices. We conclude that the weak ferromagnetic component in La_2CuIrO_6 is intrinsic which points at a noncollinear spin-structure in the ordered state.

TT 3.12 Mon 12:30 H20

Single crystal growth of α-Li_2IrO_3 from separated educts — •Friedrich Freund, Anton Jesche, Ina-Marie Pietsch, and Philipp Gegenwart — EP VI, Center for Electronic Correlations and Magnetism, Augsburg University, 86159 Augsburg, Germany

Hexagonal iridates like α-Li_2IrO_3 are promising candidates for the realization of the Kitaev exchange interaction, which describes an anisotropic and bonding dependent interaction that can lead to novel types of spin liquid behavior. We report a method to grow for the first time single crystals of α-Li_2IrO_3. This method is using separated educts and could also be used to grow single crystals of other honeycomb transition metal oxides such as Li_2RuO_3. Besides the discussion of the new crystal growth method, we will present the structural and magnetic properties.

TT 3.13 Mon 12:45 H20

Neutron scattering signatures of the 3D hyper-honeycomb Kitaev quantum spin-liquid — •Adam Smith[1], Johannes Knolle[1], Dmitry L. Kovrizhin[1,2], John T. Chalker[3], and Roderich Moessner[4] — [1]T.C.M. Group, Cavendish Laboratory, J. J. Thomson Avenue, Cambridge CB3 0HE, United Kingdom — [2]RRC Kurchatov Institute, 1 Kurchatov Square, Moscow 123182, Russia — [3]Theoretical Physics, Oxford University, 1 Keble Road, Oxford OX1 3NP, United Kingdom — [4]Max Planck Institute for the Physics of Complex Systems, D-01187 Dresden, Germany

Motivated by recent synthesis of the hyper-honeycomb material β-Li_2IrO_3, we study the dynamical structure factor (DSF) of the corresponding 3D Kitaev quantum spin-liquid (QSL), whose fractionalised degrees of freedom are Majorana fermions and emergent flux-loops. Properties of this 3D model are known to differ in important ways from those of its 2D counterpart - it has finite-temperature phase transition, as well as distinct features in Raman response. We show, however, that the qualitative behaviour of the DSF is broadly dimension-independent. Characteristics of the 3D DSF include a response gap even in the gapless QSL phase and an energy dependence deriving from the Majorana fermion density of states. Since the majority of the response is from states containing a single Majorana excitation, our results suggest inelastic neutron scattering as the spectroscopy of choice to illuminate the physics of Majorana fermions in Kitaev QSLs.