TT 47: Frustrated Magnets - Iridates and Fe-based Materials

Time: Wednesday 9:30–12:45 Location: H 0104

TT 47.1 Wed 9:30 H 0104

Dimer formation in hyperhoneycomb iridate β-Li_2IrO_3 under pressure — •Aleksandra Krajewska[1,2], Tomohiro Takayama[1,2], Alexandra Gibbs[3], Craig Bull[3], and Hidenori Takagi[1,2] — [1]1 Institut für Funktionelle Materie und Quantentechnologien, University of Stuttgart, Germany — [2]Max-Planck Institute for Solid State Research, Stuttgart, Germany — [3]ISIS Facility, Rutherford Appleton Laboratory, Didcot, UK

Honeycomb-based iridates α, β-A_2IrO_3 (A = Li, Na) are attracting attention because the magnetic coupling between $J_{\text{eff}} = 1/2$ isospins renders bond-dependent ferromagnetic interaction, providing a possible route for Kitaev quantum spin liquid (QSL). However, these materials order at low temperatures due to the presence of other magnetic couplings. β-Li_2IrO_3 shows order below 38 K. With increasing pressure, the magnetic order vanishes above 2 GPa and structural transition occurs at around 4 GPa. The driving force of the structural transition and the relevance to the QSL behaviour remains unclear. We studied the crystal structure of β-Li_2IrO_3 under pressure in detail by neutron diffraction. Above 4 GPa one of the Ir-Ir distances in the hyperhoneycomb network becomes much shorter than the others. The difference between them is 15%, similar to that in honeycomb Li_2RuO_3 with spin-singlet dimers, suggesting dimer formation in β-Li_2IrO_3. We argue that the lattice tuning by pressure modifies the balance of multiple magnetic couplings and leads to the likely destruction of $J_{\text{eff}} = 1/2$ as a consequence of dimerisation.

TT 47.2 Wed 9:45 H 0104

Coexistence of static and dynamic magnetism in pressurized β-Li_2IrO_3 — •Mayukh Majumder[1], Rudra Sekhar Manna[1], Gediminas Simutis[2], Jean-Christophe Orain[2], Tusharkanti Dey[1], Friedrich Freund[1], Rustem Khasanov[2], Pabitra Kumar Biswas[3], Alexander Tsirlin[1], and Philipp Gegenwart[1] — [1]EP-VI, EKM, University of Augsburg, Germany — [2]PSI, Villigen, Switzerland — [3]ISIS, Rutherford Appleton Laboratory, United Kingdom

Frustrated systems with Kitaev-type exchanges show long-range magnetic ordering instead of quantum spin liquid state because of the presence of other exchanges. Interestingly, theoretical studies suggest the possibility of tuning β-Li_2IrO_3 towards a QSL state by applying pressure. In order to establish the temperature-pressure phase diagram in β-Li_2IrO_3 we have used magnetization, thermal expansion, magnetostriction and μSR measurements. Our comprehensive study established - i) a weak increase of ordering temperature (T_{MO}) up to the pressure of about 14 kbar without showing any detectable change in the magnetic moment and in the nature of ordered state compared to ambient pressure, ii) above 14 kbar, T_{MO} sharply drops showing a first-order like behavior, iii) magnetic phase separation occurs above 14 kbar where static and dynamic spins coexist.

TT 47.3 Wed 10:00 H 0104

NMR studies on the single crystalline Na_2IrO_3: A model system to realize Kitaev interaction — •R. Sarkar[1], Z. Mei[2], A. Ruiz[3,4], H.-H. Klauss[1], J. G. Analytis[3,4], and N. J. Curro[2] — [1]Institute of Solid State and Materials Physics, Technical University of Dresden, 01062 Dresden, Germany — [2]Department of Physics, University of California, Davis, California 95616, USA — [3]Department of Physics, University of California, Berkeley, California 94720, USA — [4]Materials Science Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA

We present results of ^{23}Na nuclear magnetic resonance (NMR) measurements on single crystalline Na_2IrO_3, a possible candidate to realize Kitaev spin model on the honeycomb lattice. The NMR shifts ($^{23}K(\%)$), that is the measure of local susceptibility, have been studied in two crystallographic orientations. The NMR shifts reflect strong anisotropic behavior similar to the bulk susceptibility. However, below a temperature $T^* \sim 50$ K, the shift deviates from the bulk susceptibility. This anomalous behavior may be related to the exchange anisotropic bond interaction connected to the magnetic frustration. In contrast to the Knight shift, the spin-lattice relaxation rate $^{23}(1/T_1)$ is isotropic in the paramagnetic state and exhibits a strong peak at T_N. Deep in the ordered state, $^{23}(T_1T)^{-1}$ approaches a constant value as a function of temperature, suggesting the presence of significant dynamics and/or the band of excitations associated with the close proximity of quantum spin liquid ground state of Na_2IrO_3.

TT 47.4 Wed 10:15 H 0104

Tuning the Magnetism in Honeycomb Magnets — •Sebastian Selter[1], Esther Jarossay[1,2], Anja U. B. Wolter[1], Saicharan Aswartham[1], and Bernd Buechner[1,2] — [1]IFW Dresden, Dresden, Germany — [2]TU Dresden, Dresden, Germany

We present a magnetic phase diagram of $Na_2(Ir_{1-x}Pt_x)O_3$, where the magnetic order is systematically suppressed by Pt substitution. Further, preliminary data on crystal growth and magnetic behavior of $Na_2Co_2TeO_6$ and $Na_3Co_2SbO_6$ will be discussed.

Na_2IrO_3 possess a frustrated honeycomb lattice, similar to $RuCl_3$ (Ir, Ru: d^5) and $Na_2Co_2TeO_6$ and $Na_3Co_2SbO_6$ (Co: d^7). These compounds are proposed to be the best suited candidates to host a Quantum spin liquid (QSL) state. However, all these compounds show a zig-zag antiferromagnetic order at low temperatures, still indicating strong bond dependent Kitaev-type interactions. Recent experiments have shown that this magnetic order is extremely fragile. In Na_2IrO_3 already 5% of Ru substitution on the Ir site is sufficient to suppress the magnetic order, while $RuCl_3$ shows a field induced QSL state.

We address the question, how the magnetic ground state of Na_2IrO_3 is affected by dilution of the magnetic Ir honeycomb lattice with nonmagnetic Pt.

TT 47.5 Wed 10:30 H 0104

Robust spin liquid state in honeycomb iridate $D_3LiIr_2O_6$ against large isotope effect — •Tomohiro Takayama[1,2], Kentaro Kitagawa[3], Yosuke Matsumoto[1], Kenji Ishii[4], Sebastian Bette[1], Robert Dinnebier[1], and Hidenori Takagi[1,2,3] — [1]Max Planck Insitute for Solid State Research, Stuttgart, Germany — [2]University of Stuttgart, Stuttgart, Germany — [3]University of Tokyo, Tokyo, Japan — [4]QST SPring-8, Hyogo, Japan

Honeycomb iridates recently appeared as a possible materialization of Kitaev spin liquid. All hitherto-known candidate materials display a magnetic order at low temperatures likely due to the presence of additional magnetic interactions. We recently succeeded in realizing a quantum spin liquid state in proton-exchanged honeycomb iridate $H_3LiIr_2O_6$. $H_3LiIr_2O_6$ shows a large negative Weiss temperature $\theta_{\text{CW}} \sim$ -105 K, and it is not clear yet whether the Kitaev coupling is indeed a prime driving force for the spin liquid state or not.

We also synthesized an isotropic compound $D_3LiIr_2O_6$. Although $D_3LiIr_2O_6$ crystallizes basically in the same crystal structure with $H_3LiIr_2O_6$, it shows a large structural isotope effect with the elongated *c*-axis. This results in a substantial change in magnetic couplings; the antiferromagnetic couplings are enhanced as evidenced by $\theta_{\text{CW}} \sim$ -168 K. Nevertheless, $D_3LiIr_2O_6$ retains a spin-liquid state. We will discuss the differences from $H_3LiIr_2O_6$ and details of their spin-liquid state.

TT 47.6 Wed 10:45 H 0104

Synthesis and magnetic properties of Double Perovskites with Ir(IV)-states — •Michael Vogl[1], Anja Wolter[1], Sabine Wurmehl[1], Saicharan Aswartham[1], and Bernd Büchner[1,2] — [1]Leibniz-Institut für Festkörper- und Werkstoffforschung IFW Dresden — [2]Technische Universität Dresden

The magnetism of strongly spin-orbit coupled 5d-systems is a rapidly evolving topic with many open questions. In particular, Iridates are in focus for theoretical and experimental investigations.

The Double Perovskite structure, with the general notation $A_2BB'O_6$, allows the tuning of magnetic properties by doping and incorporation of various magnetic ions and sublattices. In general, Double Perovskite Iridates offer the possibility to study the magnetic exchange interaction of spin-orbit coupled Ir-ions in detail. Furthermore the B-sublattice of a Double Perovskite can be viewed as a geometrically frustrated fcc-lattice. Hence, the magnetism of these materials also touches the intriguing field of frustrated magnetism.

In this talk we present new Double Perovskite Iridates. We focus on the realization of a fcc-lattice of Ir(IV) within the Double Perovskite structure. Furthermore the interactions of Ir(IV) with rare earth ions on the A-site is examined. Particularly interesting properties were found for Nd_2ZnIrO_6. In this contribution, we present a magnetic phase diagram of this compound.

TT 47.7 Wed 11:00 H 0104

Magnetic ordering in the mixed valent iridate $Ba_3LuIr_2O_9$ — •Tusharkanti Dey[1], Mayukh Majumder[1], Jean-Christophe Orain[2], Anatoliy Senyshyn[3], Philipp Gegenwart[1], and Alexander Tsirlin[1] — [1]EP-VI, EKM, University of Augsburg, Germany — [2]Paul Scherrer Institut, Villigen PSI, Switzerland — [3]FRM II, TU Munich, Germany

Mixed valent iridates with the general formula $Ba_3MIr_2O_9$ (M is a trivalent ion) contain face-sharing Ir_2O_9 bi-octahedra with Ir-Ir dimers along the crystallographic c-axis. In these materials, Ir has a single crystallographic site with an average charge state of +4.5. The two Ir sites within the dimer share 9 electrons among them resulting in S=1/2 moments per dimer [1]. These dimers form a triangular lattice in the crystallographic ab-plane and a buckled honeycomb lattice between the neighboring planes. We have recently synthesized polycrystalline samples of $Ba_3M^{3+}Ir_2^{4.5+}O_9$ (M=In,Lu) and studied their structural and magnetic properties. In this presentation, we will discuss the results of our neutron diffraction, magnetic susceptibility, heat capacity and μSR measurements suggesting magnetically ordered ground state for $Ba_3LuIr_2O_9$ below 5.5 K and compare with the sister compound $Ba_3InIr_2O_9$ showing persistent spin dynamics and the absence of long range ordering down to 20 mK [1].
[1] T. Dey *et. al.*, Phys. Rev. B **96**, 174411 (2017)

15 min. break.

TT 47.8 Wed 11:30 H 0104

Low temperature heat capacity measurements on the mixed-valence iridate $Ba_3InIr_2O_9$ — •Sebastian Bachus, Yoshifumi Tokiwa, Tusharkanti Dey, Mayukh Majumder, Alexander Tsirlin, and Philipp Gegenwart — Experimental Physics VI, Center for Electronic Correlations and Magnetism, University of Augsburg, Germany

Frustrated magnetism can give rise to new exotic states. A prominent example are quantum spin liquids (QSL) which show no ordering of their magnetic moments even at zero temperature. Promising candidates for a QSL ground state are materials with highly frustrated magnetic moments e.g. on a kagomé or triangular lattice.
Recently the family of mixed-valence iridates has been proposed as an alternative material class for the search of QSL. One example is $Ba_3InIr_2O_9$, where mixed-valence Ir_2O_9 dimers with $Ir^{4.5+}$ are formed, sharing one unpaired electron per dimer [1]. Here, we report low temperature measurements of heat capacity. Our data show no long-range ordering as well as continuos excitations down to at least 80 mK, which strongly indicates a gapless QSL ground state. This work has been funded by the German Science Foundation through TRR 80.
[1] T. Dey *et al.*, Phys. Rev. B 96, 174411 (2017).

TT 47.9 Wed 11:45 H 0104

Magnetism in the fcc lattices of K_2IrX_6 (X=Cl, Br) crystals: Candidate $j_{\rm eff}$=1/2 Mott insulators — •Nazir Khan[1], Danil A. Prishchenko[2], Vladimir G. Mazurenko[2], Anton Jesche[1], and Alexander A. Tsirlin[1] — [1]EP VI, EKM, Augsburg University, 86159 Augsburg, Germany — [2]Ural Federal University, Mira Str. 19, 620002 Ekaterinburg, Russia

Potassium hexahaxoiridates $K_2Ir^{4+}X_6$ (X= Cl, Br) are fascinating systems to explore the physics of spin-orbit coupled $j_{\rm eff}$=1/2 moments on the geometrically frustrated fcc lattice. The structural symmetry of the system and the spin-orbit coupling in the Ir^{4+} ions allow different anisotropic exchanges besides the isotropic Heisenberg exchange ($J_{\rm H}$) such as the Kitaev ($J_{\rm K}$) and off-diagonal (J_γ) exchanges to determine its magnetic ground state. Transport measurement shows highly insulating electronic nature with charge gaps of 0.7 eV and 1.0 eV for the K_2IrCl_6 and K_2IrBr_6 compounds, respectively. Magnetization and heat capacity measurements show paramagnetic to antiferromagnetic phase transitions with Néel temperatures $T_{\rm N}$= 3.1 K and 10.2 K for the K_2IrCl_6 and K_2IrBr_6 crystals, respectively, with effective moments close to that predicted theoretically for the $j_{\rm eff}$=1/2 moment. The field dependence of the magnetization along different crystallographic directions suggests intricate nature of the exchange interactions. The structural and the magneto-electronic phase diagrams show strong dependence on the nature of the halogen ion X^- (Cl^- and Br^-). Magnetism in K_2IrCl_6 has been discussed in terms of a Heisenberg-Kitaev model.

TT 47.10 Wed 12:00 H 0104

The magnetic phase diagram and short-range magnetic order in γ-Li_2FeSiO_4 — •Martin Jonak[1], Fabian Billert[1], Sisi Gu[1,2], Christoph Neef[1], Waldemar Hergett[1], Johannes Werner[1], Changhyun Koo[1], and Rüdiger Klingeler[1] — [1]Kirchhoff Institute for Physics, University of Heidelberg, Heidelberg, Germany — [2]University of Science and Technology of China, Hefei, China

We report thermal expansion, specific heat, and high-frequency electron spin resonance (HF-ESR) studies up to 17 T, as well as pulsed-field magnetisation data on a γ-Li_2FeSiO_4 (space group $Pmnb$) single crystal. The data imply the onset of long-range antiferromagnetic order at $T_{\rm N}$ = 17.0(5) K. The uniaxial pressure dependencies are derived from our uniaxial thermal expansion data. At low temperatures, the saturation field for $B||$easy axis amounts to 33 T. In addition, there are several field-induced anomalies suggesting competing magnetic phases. The antiferromagnetic resonance modes exhibit significant zero-field splitting associated with magnetic anisotropy. At higher temperatures, the HF-ESR data imply the evolution of local magnetic fields at least up to 100 K, and are accompanied by sizeable magnetostriction well above $T_{\rm N}$.

TT 47.11 Wed 12:15 H 0104

Interplay of structure and magnetism in frustrated intermetallic AFe_4X_2 systems — Inga Kraft[1,2], Katharina Weber[1,2], Christoph Bergmann[1], Christoph Geibel[1], and •Helge Rosner[1] — [1]Max Planck Institute for Chemical Physics of Solids, Dresden — [2]Technical University of Dresden

Due to their complex and versatile behavior frustrated systems present great experimental and theoretical challenges. Even slight perturbations induce instabilities in such systems and prompt the emergence of unusual phenomena. The intermetallic AFe_4X_2 compounds (A=Sc,Y,Lu,Zr; X=Si,Ge) are suggested to cover the whole regime from frustrated AFM order up to an AFM quantum critical point. Our DFT calculations exhibit a strong interplay of structure and magnetism. However, according to our calculations, the structural and the magnetic phase transition can be considered as independent in reasonably good approximation. We discuss the influence of the A and X site atoms on the strength of magnetic interactions and the size of structural distortion.

TT 47.12 Wed 12:30 H 0104

Role of single-ion anisotropy in Fe^{3+}-based frustrated magnets — •Alexander A. Tsirlin — EP VI, EKM, University of Augsburg, Germany

Transition-metal ions with the half-filled d-shell and the high-spin d^5 electronic configuration are magnetically nearly isotropic. Here, using density-functional calculations juxtaposed with the experimental data from thermodynamic measurements and neutron diffraction, I will argue that such d^5 ions still feature a non-negligible single-ion anisotropy that prevails over intersite anisotropy terms and drives multiple magnetic transitions in frustrated magnets based on Fe^{3+}. The talk will cover Fe-based pyroxene compounds, where frustrated interchain interactions trigger a collinear spin-density-wave phase separating the ground-state helical order from the paramagnetic state. I will further report the sequence of magnetic transitions in the Cairo antiferromagnet $Bi_4Fe_5O_{13}F$, where competing single-ion anisotropies introduce two flavors of orthogonal order separated by a collinear phase. Despite their small size, typically less than 1% of the isotropic exchange couplings, the single-ion terms play pivotal role for the magnetically ordered states of Fe^{3+} compounds.