TT 55: Correlated Electrons: Low-Dimensional Systems -Materials 2

Time: Thursday 9:30–13:00 Location: H21

TT 55.1 Thu 9:30 H21

Absence of charge order in the dimerized κ-phase BEDT-TTF salts — •Martin Dressel[1], Katrin Sedlmeier[1], Sebastian Elsässer[1], David Neubauer[1], Rebecca Beyer[1], Dan Wu[1], Tomislav Ivek[1,2], Silvia Tomic[2], and John A. Schlueter[3] — [1]1. Physikalisches Institut, Universität Stuttgart, Germany — [2]Institut za fiziku, Zagreb, Croatia — [3]Material Science Division, Argonne National Laboratory, U.S.A.

Utilizing infrared vibrational spectroscopy we have investigated dimerized two-dimensional organic salts in order to search for possible charge redistribution that might constitute electronic dipoles and ferroelectricity: the quantum spin liquid κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, the antiferromagnetic Mott insulator κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl, and the superconductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br. None of them exhibit any indication of charge disproportionation. Upon cooling to low temperatures all BEDT-TTF molecules remain homogeneously charged within $\pm 0.005e$. No modification in the charge distribution is observed around $T = 6$ K where a low-temperature anomaly has been reported for the spin-liquid material κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$. In this compound the in-plane optical response and vibrational coupling are rather anisotropic, indicating that the tilt of the BEDT-TTF molecules in c-direction and their coupling to the anion layers has to be considered in the explanation of the electromagnetic properties.

[1] K. Sedlmeier *et al.*, Phys. Rev. B (in press); arXiv:1208.2652

TT 55.2 Thu 9:45 H21

Magnetoacoustic and pressure studies of frustrated distorted diamond chain compound azurite — •P. T. Cong, B. Wolf, R. S. Manna, U. Tutsch, M. de Souza, A. Brühl, and M. Lang — Physikalisches Institut, Goethe-Universität, SFB/TR 49, D-60438 Frankfurt

Measurements of ultrasound, thermal expansion and magnetic susceptibility measurements were carried out on a high-quality single crystal of distorted diamond chain compound azurite. The results show clear signatures of the magnetic energy scales involved [1, 2] and disclose two pronounced anomalies at ~ 20 K and ~ 5 K in accordance with literature results of magnetic susceptibility and specific heat data [1]. Huge anomaly of acoustic signal and thermal expansivity at $T_N = 1.88$ K demonstrate that in this material the onset of long-range antiferromagnetic (AFM) ordering is accompanied by sizable structural distortions. From the analysis of the temperature dependence of elastic constant and results of the pressure dependence of the magnetic susceptibility an estimate of the strain dependence of the dominant magnetic exchange coupling can be derived. These results highlight the significant role of strain interaction in this compound. The field and temperature dependence of sound velocity and thermal expansion reveal a very complex magnetic structure of azurite at temperature below T_N. Based on these measurements we map out a detailed $B-T$ phase diagram.

[1] H. Kikuchi et al. Phys. Rev. Lett. 94, 227201 (2005).
[2] H. Jeschke et al., Phys. Rev. Lett. 106, 217201 (2011)

TT 55.3 Thu 10:00 H21

Impurity effects in a S=1/2 Heisenberg spin chain probed by ^{63}Cu NMR — •Yannic Utz[1], Eva Maria Brüning[1], Franziska Hammerath[1], Christian Rudisch[1], Hans-Joachim Grafe[1], Ashwin Mohan[1], Romuald Saint-Martin[2], Alexandre Revcolevschi[2], Christian Hess[1], Satoshi Nishimoto[1], Stefan-Ludwig Drechsler[1], and Bernd Büchner[1] — [1]IFW Dresden, Germany — [2]LPCES, Orsay, France

We present ^{63}Cu NMR measurements on undoped, Ni doped and Mg doped $SrCuO_2$ single crystals. $SrCuO_2$ is a good realization of a one-dimensional S=1/2 Heisenberg spin chain. This is confirmed by the theoretically-expected temperature independent NMR spin-lattice relaxation rate T_1^{-1}. Doping with Ni, which can be regarded as a S=1 impurity, has a major impact on the magnetic properties of the spin chains. On the one hand, this is manifested by unusual features in the NMR spectra below 100 K, revealing the existence of an impurity-induced local alternating magnetisation. On the other hand, exponentially decacying spin lattice relaxation rates towards low temperatures indicate the opening of a spin gap similar to Ca doped $SrCuO_2$ [1]. Mg doping (S=0) has, however, no influence on the magnetic properties of the spin chains. Neither the NMR spectra nor the spin lattice relaxation rates differ from those measured on pure $SrCuO_2$. While the different impact of Ni and Mg doping on the spin chains could be explained by their different impurity spins, the opening of a spin gap in case of Ni doping is totally unexpected and not yet understood.

[1] F. Hammerath et al., Phys. Rev. Lett. 107, 017203 (2011).

TT 55.4 Thu 10:15 H21

Magnetism of CuX_2 frustrated chains (X = F, Cl, Br): the role of covalency — •Stefan Lebernegg[1], Miriam Schmitt[1], Alexander Tsirlin[1,2], Oleg Janson[1], and Helge Rosner[1] — [1]MPI Chemical Physics of Solids, Dresden, Germany — [2]National Institute of Chemical Physics and Biophysics, Tallinn, Estonia

Periodic and cluster DFT calculations, including DFT+U and hybrids, are applied to study magnetostructural correlations in spin-1/2 frustrated chain compounds: $CuCl_2$, $CuBr_2$, and a fictitious chain structure of CuF_2. The nearest-neighbor and second-neighbor exchange integrals, J_1 and J_2, are evaluated as a function of the Cu–X–Cu bridging angle θ in the physically relevant range 80–110°. The ionic CuF_2 exhibits ferromagnetic coupling only for $\theta \leq 100°$, according to the Goodenough-Kanamori-Anderson rules. However, both $CuCl_2$ and $CuBr_2$ feature ferromagnetic J_1 in the whole angular range studied. This surprising behavior is ascribed to the increased covalency in the Cl and Br compounds, which amplifies the contribution from Hund's exchange on the ligand atoms and renders J_1 ferromagnetic. At the same time, the larger spatial extent of X orbitals enhances the antiferromagnetic J_2, which is realized via the long-range Cu–X–X–Cu paths. Both, periodic and cluster approaches supply a consistent description of the magnetic behavior which is in good agreement with the experimental data for $CuCl_2$ and $CuBr_2$. Thus, owing to their simplicity, cluster calculations have excellent potential to study magnetic correlations in more involved spin lattices, especially making parameter-free quantum-chemical methods computationally feasible.

TT 55.5 Thu 10:30 H21

Novel frustrated quantum antiferromagnets in the solid-solution $Cs_2CuCl_{4-x}Br_x$through site-selective halide substitution — •Bernd Wolf, Pham Thanh Cong, Natalia van Well, Franz Ritter, Wolf Assmus, and Michael Lang — Physikalisches Institut, Goethe-Universität, SFB/TR 49, D-60438 Frankfurt

Depending on the growing conditions, an A-type orthorhombic or a B-type tetragonal structure can be found in the solid solution $Cs_2CuCl_{4-x}Br_x$ ($0 \leqslant x \leqslant 4$). Here we present measurements of the temperature-dependent susceptibility and isothermal magnetization on the B-type compounds x = 1.6 and 2.0 and compare these results with the magnetic properties recently derived for the A-type variant by Cong *et al.*, Phys. Rev. B 83, 064425 (2011). We find that due to their different Cu coordination, these two structural modifications exhibit quite dissimilar magnetic properties. The tetragonal compounds can be classified as quasi-2D ferromagnets characterized by ferromagnetic layers with a weak antiferromagnetic inter-layer coupling, whereas the orthorhombic materials, notably the border compounds x = 0 and 4, are model systems for frustrated 2D Heisenberg antiferromagnets.

TT 55.6 Thu 10:45 H21

A metal-organic spin-1/2 dimer system yielding a 2d field-induced collectively-coupled dimer phase — •U. Tutsch[1], B. Wolf[1], T. Kretz[2], H.-W. Lerner[2], M. Wagner[2], S. Wessel[3], T. Saha-Dasgupta[4], H. Jeschke[5], R. Valenti[5], and M. Lang[1] — [1]Phys. Inst., Goethe-Universität, 60438 Frankfurt — [2]Inst. f. Anorg. Chemie, Goethe-Universität, 60438 Frankfurt — [3]Inst. f. Theoret. Festkörperphysik, RWTH Aachen, 52056 Aachen — [4]S.N. Bose National Centre for Basic Sciences, Salt Lake City, Kolkata 700098, India — [5]Inst. f. Theoret. Physik, Goethe-Universität, 60438 Frankfurt

Spin-dimer systems have proven to be very useful in studying magnetic field-induced phase transitions. Here, we present high-resolution susceptibility and specific heat measurements at very low temperatures down to 0.040 K on the metal-organic spin-1/2 dimer system $C_{36}H_{48}Cu_2F_6N_8O_{12}S_2$. The *intra*-dimer coupling between the spins on the Cu sites is $J_d/k_B \approx 10$ K, making field-induced ordering accessible for standard laboratory magnets. *Ab initio* calculations suggest an effective two-dimensional (2d) structure with *inter*-dimer couplings

J_i of at least an order of magnitude smaller than J_d. This is confirmed by comparison of the experimental data with results form Quantum Monte Carlo simulations for 1d, 2d and strongly anisotropic 3d coupling schemes. We identify a region in the field vs. temperature phase diagram where the system exhibits typical 2dXY behaviour accompanied by the formation of vortices and antivortices in the effective spin configuration. The possibility of a magnetic Berezinskii-Kosterlitz-Thouless transition will be discussed.

TT 55.7 Thu 11:00 H21

Padé approximations for the magnetic susceptibilities of Heisenberg antiferromagnetic spin chains for various spin values — •Joseph Law[1] and Reinhard Kremer[2] — [1]Hochfeld-Magnetlabor Dresden, Helmholtz-Zentrum Dresden-Rossendorf, 01314 Dresden, Germany — [2]Max-Planck-Institut für Festkörperforschung, Heisenbergstr. 1, D-70569 Stuttgart, Germany

The temperature dependence of the spin susceptibilities of S=1, $\frac{3}{2}$, 2, $\frac{5}{2}$ and $\frac{7}{2}$ Heisenberg nearest-neighbor antiferromagnetic 1D spin chains was simulated via Quantum Monte Carlo calculations, within the temperature range of $0.005 \leq T^{\star} \leq 100$ and fitted to Padé approximations with deviations between the simulated and fitted data of the same order of magnitude or smaller than the Quantum Monte Carlo error. To demonstrate the practicality of our theoretical findings, we successfully compared them with the well known 1D chain compound TMMC (d^5, S=5/2) and the new S=1 Heisenberg spin chain $NiTa_2O_6$.

15 min. break

TT 55.8 Thu 11:30 H21

Similarities between the phase diagrams of the S=3/2 Heisenberg spin chains $CrXO_4$ [X=V,P] — •Joseph Law[1], Tobias Foerster[1], Robert Glaum[2], and Reinhard Kremer[3] — [1]Hochfeld-Magnetlabor Dresden, Helmholtz-Zentrum Dresden-Rossendorf, 01314 Dresden, Germany — [2]Institut für Anorganische Chemie, Universität Bonn, 53121 Bonn, Germany — [3]Max-Planck-Institut für Festkörperforschung, Heisenbergstr. 1, D-70569 Stuttgart, Germany

$CrVO_4$ and $CrPO_4$ both adopt the same structure type. They can be well described as $S=\frac{3}{2}$ Heisenberg spin chains with predominant nearest-neighbour only spin exchange interaction. We show here an in-depth investigation into the magnetic structures of both compounds which is complimented by magnetic property measurements. Wherein, we've shown that both compounds exhibit a high field phase transition into an unknown state.

TT 55.9 Thu 11:45 H21

Lattice effects in the 2D valence-bond-solid Mott insulator $EtMe_3P[Pd(dmit)_2]_2$ — •Rudra Sekhar Manna[1], Reizo Kato[2], and Michael Lang[1] — [1]Physikalisches Institut, Goethe-University Frankfurt (M), SFB/TR 49, D-60438 Frankfurt (M), Germany — [2]RIKEN, Wako, Saitama 351-0198, Japan

$EtMe_3P[Pd(dmit)_2]_2$ is a quasi-twodimensional valence-bond-solid (VBS) Mott insulator where $Pd(dmit)_2$ molecules form dimers in the conducting layer, arranged in a triangular lattice with transfer integrals t and t' ($t'/\ t = 0.87$) and an average inter-dimer exchange coupling $J/k_B \approx 250$ K. Despite this strong spin frustration the system reveals a low-temperature transition into a spin-gapped Mott insulating phase. Here we present results of the uniaxial expansion coefficients α_i on single crystals of $EtMe_3P[Pd(dmit)_2]_2$ for temperatures 1.4 K $\leq T \leq$ 200 K and magnetic fields $B \leq 8$ T. We find a sharp and well-pronounced λ-type thermal expansion anomaly at 24.1 K corresponding to the second-order phase transition into the low-T VBS phase. The discontinuity in α_i at T_{VBS} reveals a pronounced in-plane (c-axis) vs out-of-plane (b-axis) anisotropy. Strongly anisotropic expansivities also characterize the state $T > T_{VBS}$. While the in-plane c-axis shows a large positive expansivity, as often found in soft organic materials, the out-of-plane b-axis is dominated by a large negative contribution which sets in rather abruptly around 40 K. Measurements in magnetic fields up to 8 T were found to have no effect on the VBS phase transition, *i.e.*, there are no indications of a field-induced melting of the VBS state at this field level.

TT 55.10 Thu 12:00 H21

Frustrated Cairo lattice in $Bi_4Fe_5O_{13}F$ — •Alexander A. Tsirlin[1], Dmitry Batuk[2], and Artem M. Abakumov[2] — [1]National Institute for Chemical Physics and Biophysics, Tallinn, Estonia — [2]EMAT, University of Antwerp, Belgium

Cairo lattice reveals an unusual topology of frustrated magnetic interactions that form pentagonal units. We present crystal structure and magnetism of $Bi_4Fe_5O_{13}F$, a recently discovered spin-$\frac{5}{2}$ compound, which is one of the few material prototypes of the Cairo spin lattice. Thermodynamic measurements reveal a sequence of phase transitions, with the onset of magnetic order at $T_N = 178$ K and subsequent transformations at $T_1 = 62$ K and $T_2 = 71$ K. The low-temperature magnetic structure is non-collinear and commensurate, in agreement with theoretical expectations for the Cairo-lattice spin model. However, additional magnetic transitions below T_N have not been anticipated by theory. A comparative microscopic analysis of $Bi_4Fe_5O_{13}F$ and the sister compound $Bi_2Fe_4O_9$ will be presented.

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TT 55.11 Thu 12:15 H21

Magnetic-ordering transitions of the effective XY-spin-1/2 compound Cs_2CoCl_4 — •Oliver Breunig[1], Eran Sela[2], Achim Rosch[2], Benjamin Buldmann[2], Petra Becker[3], Ladislav Bohatý[3], Sandra Niesen[1], Ralf Müller[1], and Thomas Lorenz[1] — [1]II. Physikalisches Institut, Universität zu Köln — [2]Institut für Theoretische Physik, Universität zu Köln — [3]Institut für Kristallographie, Universität zu Köln

Cs_2CoCl_4 is a model system for studying the magnetism of one-dimensional spin chains with an XY-like anisotropy. It contains $CoCl_4$ tetrahedra which form chains along the crystallographic b axis. Due to a strong crystal field, an easy-plane anisotropy of magnetization is established. For symmetry reasons, easy planes in Cs_2CoCl_4 appear in two orientations. At temperatures between 0.3 and 4 K, the compound is well described by the one-dimensional XXZ model. At lower temperatures magnetic order arises due to finite inter-chain coupling. Our measurements of thermal expansion and specific heat down to 50 mK and in magnetic fields up to 3 T reveal a field-dependent ordering temperature $T_C(H)$. Depending on the orientation of the magnetic field with respect to the easy planes' orientation various ordered phases arise. We present phase diagrams for different field directions and discuss possible ordering mechanisms.

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TT 55.12 Thu 12:30 H21

Magnetic and structural properties of antiferromagnetic VF_3 — •Patrick Reuvekamp[1], Reinhard Kremer[1], Roland Eger[1], Gwilherm Nenert[2], and Thomas Hansen[2] — [1]Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany — [2]Institut Laue-Langevin, 38042 Grenoble, France

We report on a magnetic and structural investigation of layered antiferromagnetic system vanadium (III) fluoride. VF_3 crystallizes in a distorted ReO_3 structure ($R\bar{3}c$) with rotated undistorted VF_6 octahedra. The V^{+3} cations are arranged in a triangular lattice with the possibility of exhibiting magnetic frustration. Polycrystalline samples of VF_3 were investigated using heat capacity, dielectric, magnetic susceptibility and neutron powder diffraction methods. Combining our results, we confirmed that VF_3 undergoes long-range antiferromagnetic order at ∼19 K in accordance with literature [1]. The antiferromagnetic order results in a magnetic structure with the magnetic moments alternating between **a** parallel and **b** parallel alignments in the ab plane . A second phase transition can be seen at ∼120 K in the heat capacity [1] and dielectric measurements possibly associated to a minute structural distortion.

[1] A.C. Gossard, H.J. Guggenheim, F.S.L. Hsu, and R.C. Sherwood, AIP Conf. Proc., No. 5, 302 (1971).

TT 55.13 Thu 12:45 H21

Magnetic anisotropies in the Ising spin-chain compound $BaCo_2V_2O_8$ — •Sandra Niesen, Michael Seher, Gerhard Kolland, Oliver Breunig, and Thomas Lorenz — II. Physikalische Institut, Universität zu Köln

The effective Ising spin 1/2 system $BaCo_2V_2O_8$ consists of CoO_6 octahedra that form screw chains along the crystallographic c axis which are separated by nonmagnetic Ba^{2+} and V^{5+} ions. Long-range antiferromagnetic order is observed below $T_N = 5.5$ K in zero magnetic field with the spins aligned along c. Within the tetragonal ab-plane competing exchange interactions are present causing a lower symmetry of the stabilized magnetic structure. Therefore, magnetic domains occur with

ferromagnetic alignment of the spins along [100] and an antiferromagnetic one along [010], or vice versa. High resolution zero-field thermal-expansion data show, that the system undergoes a structural transition from tetragonal to orthorombic while entering the Néel phase. Here, the frustration within the *ab*-plane is lifted by a small orthorhombic splitting $(a-b)/(a+b) \neq 0$, that is below the resolution of diffraction techniques. Moreover, detailed investigations of *e.g.* magnetization, thermal expansion, specific heat, and thermal conductivity reveal an additional magnetic-field anisotropy within the *ab*-plane. The corresponding phase diagrams for magnetic fields along [100] and [110] will be presented.

This work is supported by the DFG through SFB 608.