# TT 29: Correlated Electrons: Low-dimensional Systems - Materials 2

Time: Wednesday 14:00–18:15 Location: HSZ 301

TT 29.1 Wed 14:00 HSZ 301

**Magnetic Structure and Interactions in the Quasi-1D Antiferromagnet $CaV_2O_4$** — •Oliver Pieper[1,2], Bella Lake[1,2], Aziz Daoud-Aladine[3], Manfred Reehuis[1,4], Karel Prokeš[1], Bastian Klemke[1], Klaus Kiefer[1], Jiaqiang Yan[5], Asad Niazi[5], David C. Johnston[5], and Andreas Honecker[6] — [1]Helmholtz-Zentrum Berlin für Materialien und Energie, Berlin, Germany — [2]Technische Universität Berlin, Institut für Festkörperphysik, Berlin, Germany — [3]ISIS Facility, Rutherford Appleton Lab, Chilton, UK — [4]Max-Planck-Institut für Festkörperforschung, Stuttgart, Germany — [5]Ames Lab. and Departement of Physics and Astronomy, Iowa State University, Ames, USA — [6]Universität Göttingen, Institut für theoretische Physik, Göttingen, Germany

$CaV_2O_4$ is a spin-1 antiferromagnet where the magnetic vanadium ions are arranged on quasi-one-dimensional zig-zag chains with frustrated antiferromagnetic exchange interactions. Here we present high temperature susceptibility and single-crystal neutron diffraction measurements, which are used to deduce the magnetic structure, dominant exchange interactions and orbital configurations. The results suggest that at high temperatures of $CaV_2O_4$, the zig-zags behave as Haldane chains but at low temperatures, orbital ordering lifts the exchange frustration and the zig-zags become spin-1 ladders.

TT 29.2 Wed 14:15 HSZ 301

**Strongly dispersive magnetic excitations in $Li_2CuO_2$** — •W.E.A. Lorenz[1], S.-L. Drechsler[1], R. Kuzian[3], W. D. Stein[2], N. Wizent[1], A. Hiess[4], W. Schmidt[5], R. Klingeler[1], M. Loewenhaupt[2], and B. Büchner[1] — [1]Leibniz Institute for Solid State and Materials Research (IFW) Dresden, Germany — [2]Institut für Festkörperphysik, TU Dresden, Germany — [3]Institute for Problems of Materials Science Krzhizhanovskogo 3, 03180 Kiev, Ukraine — [4]Institut Laue Langevin, F-38042 Grenoble Cedex 9, France — [5]Jülich Centre for Neutron Science JCNS, Jülich, Germany

We report on recent inelastic neutron scattering experiments on $Li_2CuO_2$ which is considered to be the prototypical ferromagnetic $S = \frac{1}{2}$ spin chain compound. Although $Li_2CuO_2$ has been subject to many experimental and theoretical studies, its magnetic dimensionality is still discussed controversially. This open question is resolved by our spectroscopic study providing a set of magnetic exchange parameters. We find that the magnon excitation is strongly dispersive only for moment transfer along the chains. Analysis of our data by means of linear spin wave theory provides unequivocal evidence that the compound is indeed a quasi-one-dimensional magnet. To be more specific, we find that the ferromagnetic nearest neighbor intra-chain coupling is the dominant magnetic interaction, but highly frustrated by competing antiferromagnetic next-nearest neighbor couplings. In comparison, the inter-chain exchange is relatively weak. Some of the magnetic properties of the material remain puzzling and will have to be discussed with respect to highly frustrated magnetism.

TT 29.3 Wed 14:30 HSZ 301

**Ultrasonic investigations on the 1D diamond chain compound Azurite** — •Cong T. Pham[1], Andreas Brühl[1], Mariano de Souza[1], Bernd Wolf[1], Jürgen Schreuer[2], Stefan Süllow[3], and Michael Lang[1] — [1]Physikalisches Institut, Universität Frankfurt, SFB/TR49, 60438 Frankfurt(M — [2]Institut für Mineralogie, Ruhr-Universität, 44780 Bochum — [3]Institut für Physik der kondensierten Materie, TU Braunschweig, 38106 Braunschweig

The natural mineral Azurite has been considered a model substance for the 1D distorted antiferromagnetic (AFM) diamond chain. The unusual magnetic properties were believed to arise from the competition between quantum fluctuations and spin frustration and have been modelled by assuming three AFM exchange-coupling constant J1,J2,J3 > 0. However, the microscopic magnetic structure and the detailed phase diagram are unknown or disputed. Recent inelastic neutron scattering experiments suggest a non-frustrated structure with J3 < 0 and J1 ˜ 0 below the Néel-temperature of 1.8 K. In this contribution, we present results of ultrasonic investigations on a high-quality single crystal. The longitudinal elastic constant has been studied as a function of temperature down to 80 mK and magnetic fields up to 12 Tesla. The data reveal clear signatures of the magnetic energy scales involved and disclose distinct anomalies at the magnetic phase transition. Based on these results a detailed B-T phase diagram is obtained which includes an additional phase boundary of unknown origin at low temperature (T < 0.6 K). The latter is accompanied by a pronounced softening of the elastic constant.

TT 29.4 Wed 14:45 HSZ 301

**Quantized Spin Waves in a Mesoscopic Antiferromagnetic Molecular Ring** — •J. Dreiser[1], G. Carver[2], C. Dobe[2], H. U. Güdel[2], A. L. Barra[3], J. Taylor[4], and O. Waldmann[1] — [1]Physikalisches Institut, Universität Freiburg, D-79104 Freiburg, Germany — [2]Department of Chemistry and Biochemistry, University of Bern, 3012 Bern, Switzerland — [3]Grenoble High Magnetic Field Laboratory, CNRS, 38042 Grenoble Cedex 9, France — [4]ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot, Oxfordshire OX11 0QX, United Kingdom

We present inelastic neutron scattering (INS) data as well as numerical simulations on the antiferromagnetic (AF) $CsFe_8$ molecule, in which eight spin-5/2 Fe(III) ions are arranged in a ring with nearest-neighbor AF Heisenberg interaction. In such even-numbered rings the lowest-lying excitations are formed by rotational modes of the Néel vector ($L$ Band) while spin waves ($E$ Band) occur at higher energies. This is in contrast to bulk antiferromagnets where spin waves are the lowest observed excitations. Further, spin waves become quantized, i.e., occur at discrete energies due to the mesoscopic size of the ring.

We recorded INS data up to high energies at the time-of-flight spectrometer MARI at ISIS, which clearly allowed us to observe the excitation of discrete spin waves. The data is modeled using a Heisenberg-exchange Hamiltonian together with a single-ion anisotropy term. Due to the molecules' symmetry, only two parameters $J$, $D$ are needed and exact numerical diagonalization yields excellent agreement with the data.

TT 29.5 Wed 15:00 HSZ 301

**Quantized Spin Waves in a Ferromagnetic Supertetrahedron: INS studies on $Mn_{10}$** — •S. Stuiber[1], O. Waldmann[1], Y. H. Lan[2], N. Ghenadie[2], A. K. Powell[2], and T. Unruh[3] — [1]Physikalisches Institut, Universität Freiburg, D-79104 Freiburg, Germany — [2]Institut für Anorganische Chemie, Universität Karlsruhe, D-76128 Karlsruhe, Germany — [3]Technische Universität München, Forschungsneutronenquelle FRM-II, D-85747 Garching, Germany

Similar to extended systems also in a ferromagnetic cluster a spin-wave approach can be used to describe the low-lying magnetic excitations. In a cluster, however, it leads to quantized spin waves with discrete energies. In this work, we report inelastic neutron scattering (INS) experiments on a $Mn_{10}$ cluster, performed at the direct time-of-flight spectrometer TOFTOF (FRM-II). This molecule is comprised of an octahedron of $Mn^{3+}$ ions enclosed by a tetrahedron of $Mn^{2+}$ ions, forming a supertetrahedron. Ferromagnetic couplings between the Mn ions lead to a ground state with total spin $S = 22$. The spin waves then correspond to the states in the $S = 21$ sector. INS is the experiment of choice to investigate this kind of excitations, because of its favorable selection rules. In our experiment we observed these excitations and used spin-wave calculations as well as exact diagonalization studies to model the data and infer values for the exchange coupling constants.

**15 min. break**

TT 29.6 Wed 15:30 HSZ 301

**Fluctuation spectroscopy studies of the quasi–2D organic conductors $\kappa-(BEDT-TTF)_2X$** — •Jens Brandenburg[1], Jens Müller[1], and John A. Schlueter[2] — [1]Max Planck Insitute of Chemical Physics of Solids, Dresden, Saxony, Germany — [2]Argonne National Laboratory, Argonne, Illinois, USA

The family of quasi–2D organic conductors $\kappa-(BEDT-TTF)_2X$ with X = Cu[N(CN)$_2$]Cl, Cu[N(CN)$_2$]Br and Cu(NCS)$_2$ are model systems for low–dimensional metals exhibiting both strong electron–electron and electron–phonon interactions. The interplay of charge, spin, and lattice degrees of freedom lead to a variety of different magnetic–insulating, metallic, and superconducting ground states [1]. Fluctuation spectroscopy has been used to study the intrinsic charge carrier dynamics in this class of materials for the first time. Here, noise measurements as a function of temperature and magnetic field are pre-

sented for three compounds located at different positions of the phase diagram. A comparison with the resistance of the samples reveals additional features one of which can be assigned to the activation energy of the glass–like ordering of the ethylene endgroups of the ET–molecules [2]. For the materials close to the bandwidth controlled Mott transition we discuss a steep increase of the noise power upon decreasing temperature in the framework of percolation theory as due to the coexistence of antiferromagnetic insulating and superconducting phases [2].

[1] N. Toyota, M. Lang, J. Müller, Low–Dimensional Molecular Metals, Springer (2007).

[2] J. Müller, J. Brandenburg, J. A. Schlueter; to be published

TT 29.7 Wed 15:45 HSZ 301

**Resistivity investigations on the X-ray irradiated Mott insulator $\kappa$-(ET)$_2$Cu[N(CN)$_2$]Cl** — •Ammar Naji[1], Ulrich Tutsch[1], Michael Lang[1], and Takahiko Sasaki[2] — [1]Physikalisches Institut, Goethe-Universität Frankfurt (M), SFB/TRR49, D-60438 Frankfurt am Main — [2]Institute for Material Research, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai, Miyagi 980-8577, Japan

The compound $\kappa$-(ET)$_2$Cu[N(CN)$_2$]Cl is a two- dimensionally structured organic material with a rich pressure vs. temperature phase diagram including paramagnetic and antiferromagnetic Mott-insulating states as well as metallic, semiconducting and superconducting phases, see e.g., [1]. We used X-ray irradiation to induce defects in the material and by this change the charge distribution between anion- and ET-layers. The smaller resistivity of the irradiated samples compared to non-irradiated ones supports this idea [2]. As a consequence, X-ray irradiation may provide a means for doping the Mott-insulator away from half-filling. We will present resistivity data for the temperature range 5 K $\leq$ T $\leq$ 60K and pressures up to 50 MPa from which the p-T-phase diagram of X-ray irradiated $\kappa$-(ET)$_2$Cu[N(CN)$_2$]Cl will be extracted.

[1] S. Lefebvre *et al.*, Phys. Rev. Lett. **85**, 5420 (2000)

[2] T. Sasaki *et al.*, J. Phys. Soc. Jpn. **76**, 123701 (2007)

TT 29.8 Wed 16:00 HSZ 301

**Thin films of charge transfer compounds of tetrathiafulvalene and its derivatives** — •Vita Levitan and Michael Huth — Physikalisches Institut, Goethe-Universität, Frankfurt am Main, Germany

Thin films of the organic C(harge) T(ransfer) complex ET-TCNQ have been prepared by molecular beam epitaxy (MBE). We studied the growth of this compound by co-deposition and sequential deposition of two layers of ET and TCNQ molecules followed by a post growth annealing. At the layers' interface ET-TCNQ has been detected and characterized by x-ray diffraction and energy dispersive x-ray spectroscopy as the monoclinic phase of ET-TCNQ. The electrical properties of the bilayer (such as conductivity) were measured and analyzed.

We also prepared thin films of TTF-TCNQ with a preferred orientation of the microcrystals on the substrate along the b-axis of TTF-TCNQ, being the highest conductivity axis of the compound. Various oxide-based substrate materials with orientations optimized for small layer-substrate lattice mismatch were chosen. We studied the influence of epitaxial clamping on the Peierls transition of TTF-TCNQ at about 60 K by means of temperature dependence transport measurement.

(TTF: tetrathiafulvalene, ET: bis(ethylenedithio)tetrathiafulvalene, TCNQ: tetracyanoquinodimethane)

TT 29.9 Wed 16:15 HSZ 301

**Thermodynamic studies on the proposed 2D spin-liquid state in $\kappa$-(ET)$_2$Cu$_2$(CN)$_3$** — •Rudra Sekhar Manna[1], Andreas Bruehl[1], Mariano de Souza[1], John A. Schlueter[2], and Michael Lang[1] — [1]Physikalisches Institut, J.W. Goethe-Universitaet Frankfurt, SFB/TR49, D-60438 Frankfurt(M), Germany — [2]Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439, USA

The charge-transfer salt $\kappa$-(ET)$_2$Cu$_2$(CN)$_3$, where ET = BEDT-TTF, is a spin S = 1/2 Mott insulator with a twodimensional (2D) triangular lattice structure. Because of the almost perfect geometrical frustration $t'$/t $\approx$ 1 in this material, it has been proposed that quantum fluctuations destabilize long-range magnetic order and, instead, lead to liquid-like properties of the spin system [1]. Issues of high current interest include the nature of the low-lying elementary excitations of the spin liquid, particularly whether or not there is a spin gap, as well as the character of the anomaly observed around 6 K. The latter has been attributed to some "crossover"or "hidden order"[2]. Here we report on high-resolution thermal expansion and specific heat measurements focusing on the mysterious 6 K anomaly. We observe that this anomaly is accompanied by distinct and sharp features in the uniaxial-expansion coefficients suggestive of a second-order phase transition rather than a crossover. The data reveal a pronounced in-plane anisotropy corresponding to a significant distortion of the isotropic triangular lattice.

[1] Shimizu et al., Phys. Rev. Lett. 91, 107001 (2003).

[2] Yamashita et al., Nature Phys. 4, 459 (2008).

TT 29.10 Wed 16:30 HSZ 301

**Spin-state polaron in lightly hole doped $LaCoO_3$** — •A. Alfonsov[1], E. Vavilova[1,2], V. Kataev[1], B. Büchner[1], A. Podlesnyak[3], M. Russina[3], A. Furrer[4], Th. Strässle[4], E. Pomjakushina[4,5], K. Conder[5], and D.I. Khomskii[6] — [1]IFW Dresden, D-01171 Dresden, Germany — [2]Zavoisky Physical Technical Institute, RAS, 420029 Kazan, Russia — [3]Hahn–Meitner–Institut, D-14109 Berlin, Germany — [4]Laboratory for Neutron Scattering, ETH Zürich & PSI, CH-5232 Villigen PSI, Switzerland — [5]Laboratory for Developments and Methods, PSI, CH-5232 Villigen PSI, Switzerland — [6]II. Physikalisches Institut, Universität zu Köln, 50937 Köln, Germany

$LaCoO_3$ is nonmagnetic at low temperatures and shows a $T$-activated magnetism due to a change of the $Co^{3+}$ spin state. Surprisingly, a very small $Sr^{2+}$ (i.e. hole) doping yields a strong magnetic response already at low $T$. To establish its nature we performed electron spin- (ESR), nuclear magnetic resonance (NMR) and inelastic neutron scattering (INS) measurements of lightly doped $La_{1-x}Sr_xCoO_3$ single crystals ($x \sim 0.002$). $^{59}$Co NMR data indicate the formation of extended magnetic clusters distributed over the entire sample volume. Low-$T$ ESR spectra show up multiple lines with large g-factors suggesting that these clusters have a large spin multiplicity with a strong orbital contribution. From the Q-dependence of the INS intensity we conclude that the cluster comprises 7 magnetic Co ions. We propose a mechanism of how already a very light hole doping yields a formation of big spin polarons in $LaCoO_3$.

**15 min. break**

TT 29.11 Wed 17:00 HSZ 301

**Anomalous dielectric response of the copper oxide chains in $LiCuVO_4$.** — •Yulia Matiks, Peter Horsch, Reinhard Kremer, Bernhard Keimer, and Alexander Boris — Max-Planck-Institut für Festkörperforschung, 70569 Stuttgart

Spectroscopic ellipsometry has been used to investigate the low-lying electronic excitations in the strongly correlated chain copper oxide $LiCuVO_4$ in the range 0.7 - 6.5 eV at temperatures 6 - 350 K. The Zhang-Rice singlet (ZRS) state has been evidenced in edge-sharing copper oxide chains for the first time. Temperature dependence of the ZRS peak identified in the dielectric response along the chains follows spin correlations within the chains that make ZRS apparent at 6 K but strongly suppressed and invisible at 300 K. This behavior is in a good agreement with recent cluster calculations within a pd-Hubbard model. [1] The integrated optical conductivity for polarization along the chains confirms the redistribution of the spectral weight between ZRS and non-bonding O2p states below 80 K. The corresponding effective number of charge carriers transferred between the states is 0.02 electrons per Cu atom indicating the kinetic energy gain of about 10 meV, which is of the same order of magnitude as the exchange integrals of the competing nearest $J_1$ = -2.1 meV and next-nearest $J_2$ = 4.3 meV neighbor interactions [2]. For polarization perpendicular to the chains, no spectral weight transfer between individual bands is observed.

[1] J. Málek et al., Phys. Rev. B 78, 060508 (R) (2008).

[2] M. Enderle et al., Europhys. Lett. 70, 237 (2005).

TT 29.12 Wed 17:15 HSZ 301

**End states and singlet-triplet degeneracy in linear atomic chains** — Antonio Monari[1], Valentina Vetere[2], Gian Luigi Bandazzoli[1], Stefano Evangelisti[2], and •Beate Paulus[3] — [1]Dipartimento di Chimica Fisica e Inorganica, Universita di Bologna, Bologna, Italy — [2]Laboratoire de Chimie et Physique Quantiques, Universite de Toulouse et CNRS, Toulouse Cedex, France — [3]Institut für Chemie und Biochemie, Frei Universität Berlin, 14195 Berlin

The electronic structure of linear beryllium chains has been theoretically studied by using ab initio methods. It turns out that, for internuclear distances close to the equilibrium structure, two partially

filled edge orbitals ('edge states') localize at the chain ends. This gives rise to two low-lying states, a singlet ground state, $^1\Sigma_g$ and a quasi-degenerate triplet, $^3\Sigma_u$. the energy splitting goes rapidly to zero as the number of atoms in the chain is increased. Preliminary investigations indicate that this could be a general behavior shard by metals belonging to the groups 2 and 12.

TT 29.13 Wed 17:30 HSZ 301

**Understanding magnetic properties of layered copper oxychloride (CuCl)$LaNb_2O_7$** — •Alexander Tsirlin[1,2] and Helge Rosner[1] — [1]Max-Planck Institute CPfS, Dresden, Germany — [2]Department of Chemistry, MSU, Moscow, Russia

The copper-containing oxychloride (CuCl)$LaNb_2O_7$ is one of the actively studied low-dimensional spin systems. Experimental data evidence the strong frustration of this compound, but the microscopic scenario of the frustration remains unknown. Initially, (CuCl)$LaNb_2O_7$ was considered as a promising realization of the spin-1/2 frustrated square lattice model. However, further studies suggested the sizable structural distortion, leading to a more complex spin physics. Experimental techniques fail to resolve the distorted structure, hence impeding the understanding of the magnetic properties. In this contribution, we present a computational study of (CuCl)$LaNb_2O_7$ and propose a valid miscroscopic scenario that accounts for all the experimental data available so far. Our results indicate orbital degeneracy as a primary origin of the structural distortion. The distorted structure is orthorhombic and includes $CuO_2Cl_2$ plaquettes instead of $CuO_2Cl_4$ octahedra in the regular structure. The distortion strongly modifies the exchange couplings in (CuCl)$LaNb_2O_7$ and leads to a three-dimensional spin model with pronounced one-dimensional features. Our findings propose a general scenario for the structural distortion and the spin physics of (CuX)$LaM_2O_7$ (X = Cl, Br; M = Nb, Ta) as well as an outlook for further experimental studies of these compounds.

TT 29.14 Wed 17:45 HSZ 301

**Volborthite $Cu_3V_2O_7(OH)_2\cdot 2H_2O$: Orbital ordering on a distorted kagome geometry** — •Oleg Janson[1], Johannes Richter[2], and Helge Rosner[1] — [1]MPI CPfS Dresden, Germany — [2]University of Magdeburg

The mineral volborthite $Cu_3V_2O_7(OH)_2\cdot 2H_2O$ has been recently claimed as a candidate for a real material realization of a spin-1/2 kagomé lattice. Despite the monoclinic distortion which allows non-frustrated magnetic couplings, experiments reveal the presence of frustration and no clear evidence for long-range order down to 50 mK. Here, we present a theoretical investigation of this interesting material. Based on DFT calculations, we estimate the relevant orbitals and couplings. Quite unusual for cuprates, we find different magnetically active orbitals for structurally different Cu(I) and Cu(II) atoms: while the latter has a hole in a $x^2 - y^2$ orbital (like in most cuprates), for Cu(I) the $3z^2 - r^2$ is half-filled like in $CuSb_2O_6$. This has a dramatic influence on exchange integrals, which have been estimated using the LSDA+$U$ approach. The magnetic degrees of freedom are mapped onto a Heisenberg model. We find that the orbital order leads to a picture of coupled frustrated chains rather than a kagomé model. For the proposed spin model we investigate the classical as well the quantum ground state. The possible influence of spin anisotropy and Dzyaloshinskii-Moriya interactions will be briefly discussed.
ZIH and IFW Dresden are acknowledged for support with computational resources.

TT 29.15 Wed 18:00 HSZ 301

**Quantum phase transitions in $Ca_3Ru_2O_7$** — •Oliver Welzel[1], Patricia Alireza[1], Carsten Albrecht[2], Naoki Kikugawa[3], Andrew Mackenzie[3], and Malte Grosche[1] — [1]Cavendish Laboratory, Cambridge, UK — [2]Royal Holloway, University of London, Egham, UK — [3]School of Physics and Astronomy, University of St. Andrews, UK

The strongly correlated bilayer ruthenate $Ca_3Ru_2O_7$ undergoes first a magnetic transition ($T_N = 56$ K, $\mathbf{Q} = (0, 0, 1/2)$) and then a structural transition ($T_S = 48$ K) on cooling. At low temperature, this material exhibits very small Fermi surface pockets, consistent with its low carrier density.

We follow the evolution of the low temperature state of $Ca_3Ru_2O_7$ with pressure: The structural transition is rapidly suppressed, and $T_S$ extrapolates to zero at $p_{c1} \simeq 35$ kbar. The magnetic transition is also suppressed. $T_N$ extrapolates towards a critical pressure of $p_{c2} \simeq 55$ kbar, but above 40 kbar it is replaced by signatures of a new ordered phase with an onset temperature of about $T_X \simeq 25$ K. This new ordered phase, in turn, appears to be suppressed near $p_{c3} \simeq 80$ kbar.

We explore the nature of the high pressure state of $Ca_3Ru_2O_7$ in a series of high pressure measurements of the resistivity, magnetic susceptibility and magnetisation, and investigate, in particular, the vicinity of the quantum critical point, $p_{c3}$.