TT 13: Correlated Electrons: Low-dimensional Systems - Materials 1

Time: Tuesday 9:30–13:00 Location: H 2053

TT 13.1 Tue 9:30 H 2053

Resonant inelastic soft x-ray scattering on the spin-ladder/spin-chain system $Sr_{14}Cu_{24}O_{41}$ — •Justina Schlappa[1], T. Schmitt[1], A. Bendounan[1], X. Wang[2], A. Piazzalunga[3], V. Strocov[1], B. Delley[1], B. Thielemann[1,4], H. Ronnow[2], G. Ghiringhelli[3], M. Grioni[3], L. Braicovich[3], C. Dallera[3], J. Mesot[1,4], and L. Patthey[1] — [1]Paul Scherrer Institut, Switzerland — [2]Ecole Polytechnique Fédérale Lausanne, Switzerland — [3]Politecnico di Milano, Italy — [4]ETH Zürich, Switzerland

The layered system $(Sr,Ca)_{14}Cu_{24}O_{41}$, is a low-dimensional cuprate system that has been studied recently with large interest [1]. It is mixed-valent and built up of two different cupper-oxygen layers: 1-dim CuO_2 chains and quasi 1-dim Cu_2O_3 2-leg ladders. When cooling below 200 K a full range of interresting phenomena is observed, as structural changes, formation of charge order and formation of AF-dimers on the chains.

We investigated the electronic structure of $Sr_{14}Cu_{24}O_{41}$ by inelastic x-ray scattering (RIXS) [2] at the Cu $M_{2,3}$ edge ($3p \rightarrow 3d$ transition). The spectra show well-pronounced inelastic signal in the constant energy loss of 1.5-2.5 eV, which originates from crystal field excitations. Differences between the different crystallographic directions and between the high- and low-temperature phase are revealed.

[1] T. Vuletic et al., Physics Reports 428, 169 (2006).

[2] A. Kotani and S. Shin, Rev. Mod. Phys. 73, 203 (2001).

TT 13.2 Tue 9:45 H 2053

Highly frustrated $S = 1/2$ spin chains near a quantum critical point — •R. Klingeler[1], S.-L. Drechsler[1], N. Tristan[1], V. Kataev[1], F. Kretzschmar[1], Y. Arango[1], N. Leps[1], J. Vavilova[1], A. Parameswaran[1], H.-H. Klauss[2], H. Luetkens[3], O. Volkova[4], A. Vasiliev[4], T. Lorenz[5], H. Rakoto[6], U. Zeitler[7], J. Richter[8], and B. Büchner[1] — [1]Leibniz Institute for Solid State and Materials Research (IFW) Dresden — [2]TU Dresden — [3]PSI Villingen — [4]Moscow State University — [5]University of Köln — [6]LNCMP Toulouse — [7]HMFL Nijmegen — [8]University Magdeburg

Frustrated antiferromagnetic quantum spin chains with competing nearest and next-nearest neighbor interactions exhibit a rich physics with unusual ground states. Effects of quantum fluctuations are particularly strong in the vicinity of a quantum critical point where small perturbations such as a weak external magnetic field or the inter-chain coupling strongly affect the physical properties. This situation is realized in the novel S=1/2 spin chain compound Li_2ZrCuO_4 which is close to a quantum critical point. We report on specific heat, thermal expansion and magnetisation as well as on NMR, ESR and μSR studies which confirm high frustration and strong quantum fluctuations. While inter-chain coupling yields long range antiferromagnetic order at low temperatures our data suggest short range helical/AFM correlations above T_N. Upon application of a magnetic field of 9T, however, these correlations are suppressed against ferromagnetic ones. ESR and NMR data confirm the coexistence of quasi-1D behavior and short range AFM correlations up to $80\,K \gg T_N \approx 7\,K$.

TT 13.3 Tue 10:00 H 2053

Low-dimensional spin-1/2 systems in complex vanadium phosphates — •Alexander Tsirlin[1,2], Ramesh Nath[1], Christoph Geibel[1], and Helge Rosner[1] — [1]Max-Planck Institute CPfS, Dresden, Germany — [2]Department of Chemistry, MSU, Moscow, Russia

Vanadium phosphates are known to reveal low-dimensional spin-1/2 systems and to provide unique realizations for some of the actively studied spin models like frustrated J_1-J_2 square lattice. Magnetic patterns of these compounds may be quite complicated as non-magnetic phosphate tetrahedra mediate superexchange interactions. The signs and the magnitudes of such interactions can hardly be estimated on structural basis only, therefore the choice of the appropriate spin model is sometimes very problematic. In our contribution we present the results of joint experimental (thermodynamic properties measurements) and computational studies of several complex vanadium phosphates. We employ full-potential band structure calculations in order to give reliable estimates of the exchange coupling constants. The estimates justify the choice of the particular model and help us to give reliable interpretation of the experimental data. We reach a remarkable agreement between the experimental and computational results. Our studies show that some of the vanadium phosphates are promising objects for the investigation of low-temperature quantum phenomena in low-dimensional spin-1/2 systems.

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TT 13.4 Tue 10:15 H 2053

$KTi(SO_4)_2.H_2O$ - a possible candidate for a new spin-Peirles system — •Deepa Kasinathan[1], Goran Nilsen[2], Henrik Ronnow[2], Stefan-Ludwig Drechsler[3], and Helge Rosner[1] — [1]MPI CPfS - Dresden, Germany — [2]LQM-EPFL, Lausanne, Switzerland — [3]IFW Dresden, Germany

Recently a large number of compounds belonging to the family of $J_1 - J_2$ chain models with competing ferromagnetic (FM) and antiferromagnetic (AFM) interactions have been discovered. In most cases, FM-J_1 and AFM-J_2 is observed, leading to helical order with no spin gap (for frustration ratio $\alpha = \frac{J_1}{J_2} >$ -0.25). Systems with both J_1 and J_2 being AFM causing a spin gap are rather rare. The thermodynamic data of the recently prepared $KTi(SO_4)_2.H_2O$ reveal that this system is a quasi 1D spin 1/2 chain compound with both J_1 and J_2 being AFM, and a frustration ratio $\alpha \approx 0.29$. Here we report the results of electronic structure calculations within the LSDA+U method along with tight-binding models. Our calculations confirm that both J_1 and J_2 are AFM. In contrast to the experiments we obtain a larger α, slightly depending on the choice of the Coulomb repulsion U. Therefore $KTi(SO_4)_2.H_2O$ might be a new candidate for a spin-Peirles ground state. A brief comparison with other systems belonging to the class of frustrated chain materials is given with respect to their position in the general phase diagram of the 1D $J_1 - J_2$ model.

TT 13.5 Tue 10:30 H 2053

Spin dynamics in the plateau phase of the 1D distorted diamond chain azurite, $Cu_3(CO_3)_2(OH)_2$ — Kirrily Rule[1], Anja Wolter[1], •Stefan Süllow[2], Bernd Wolf[3], Michael Lang[3], Jürgen Schreuer[4], and Alan Tennant[1] — [1]BENSC, HMI, 14109 Berlin — [2]IPKM, TU Braunschweig, 38106 Braunschweig — [3]Physikalisches Institut, J.W. Goethe-Universität Frankfurt, 60438 Frankfurt(M) — [4]Ruhr-Universität Bochum, Bochum

Recently, azurite, $Cu_3(CO_3)_2(OH)_2$, has been described as an ideal example of a frustrated one-dimensional (1D) diamond-chain antiferromagnet [1]. A 1/3 magnetisation plateau has been observed in azurite for fields between 11 and 30T applied perpendicular to both the crystallographic b axis and Cu^{2+} chain direction. Here, we present a study by means of inelastic neutron scattering, which has been performed on a large single crystal of azurite in fields up to 14T applied perpendicular to the chain direction, *viz.* in the plateau phase. Our data in the plateau phase indicate that the magnetic excitations can be described by a dimer-monomer model with two distinct energy scales. The dispersion function of the monomer chain reveals an effective spin coupling of $J_{eff} = 10.1(2)$ K, while the lowest Zeeman-split dimer branch has an effective dimer-dimer coupling of $J_{dimer} = 1.8(1)$ K. We will discuss possible values for the exchange constants J_1, J_2 and J_3 of the diamond chain model to account for these observations, based on a perturbative approach.

[1] H. Kikuchi et al. Phys. Rev. Lett. **94** (2005) 227201

TT 13.6 Tue 10:45 H 2053

On the microscopic description of the spin-1/2 quantum magnet azurite — •Andreas Honecker[1], Johannes Richter[2], Helge Rosner[3], Oleg Janson[3], Roser Valenti[4], Hem Kandpal[4], Hena Das[5], Bernd Wolf[6], and Michael Lang[6] — [1]Institut für Theoretische Physik, Georg-August-Universität Göttingen — [2]Institut für Theoretische Physik, Otto-von-Guericke Universität Magdeburg — [3]Max-Planck-Institut für Chemische Physik fester Stoffe, Dresden — [4]Institut für Theoretische Physik, J.W. Goethe-Universität Frankfurt — [5]S.N. Bose National Centre for Basic Sciences, Kolkata, India — [6]Physikalisches Institut, J.W. Goethe-Universität Frankfurt

The mineral azurite $Cu_3(CO_3)_2(OH)_2$ exhibits interesting low-temperature properties like a magnetization plateau at 1/3 of saturation [H. Kikuchi *et al.*, Phys. Rev. Lett. **94** (2005) 227201]. While the crystal structure suggests a description in terms of the spin-1/2 Heisenberg model on a distorted diamond chain, the values of the associated ex-

change constants have remained a matter of debate. Firstly, we present a study of the complete parameter space of the spin-1/2 Heisenberg model on the distorted diamond chain. Thermodynamic and dynamic properties are computed by exact diagonalization and compared to experimental results for azurite. In particular, we discuss the constraints obtained from recent inelastic neutron scattering results on the 1/3 plateau [K.C. Rule *et al.*, arXiv:0709.2560]. Secondly, complementary *ab-initio* computations are performed in order to obtain further information about the relevant exchange paths in azurite as well as the values of the associated magnetic exchange constants.

TT 13.7 Tue 11:00 H 2053

Molecule-based realization of an $S = 1/2$ antiferromagnetic Heisenberg chain — •K. Remović-Langer[1], Y. Tsui[1], U. Tutsch[1], B. Wolf[1], W. Assmus[1], A. Prokofiev[4], A. Honecker[2], G. Donath[3], and M. Lang[1] — [1]Physikalisches Institut, J.W. Goethe-Universität, Max-von-Laue-Str. 1, SFB/TR 49, D-60438 Frankfurt(M). — [2]Institut für Theoretische Physik, Georg-August-Universität Göttingen, D-37077 Göttingen. — [3]Max-Planck-Institut für Chemische Physik fester Stoffe, Nöthnitzer Str. 40, D-01187 Dresden. — [4]Institut für Festkörperphysik, Technische Universität Wien, Wiedner Hauptstr. 8-10, A-1040 Wien.

Generally, fine tuning of an external control parameter is required to drive a system to a quantum critical point (QCP). This is different for the spin $S = 1/2$ antiferromagnetic Heisenberg chain (AFHC) which is inherently quantum critical. Here we present magnetic and thermodynamic measurements on single crystals of a copper coordination polymer $[Cu(\mu\text{-ox})(4\text{-apy})_2(H_2O)]_n$. Measurements of the susceptibility, magnetization and specific heat were found to be in full accordance with theoretical predictions for the uniform $S = 1/2$ AFHC with a small magnetic exchange interaction of $J/k_B \sim (3.1\pm0.1)$K. The material can be easily tuned to the saturation field $B_s \sim 4.2$ T, given by $g\mu_B B_s = 2J$, which marks the endpoint of a quantum critical line in the $T - B$ plane.

15 min. break

TT 13.8 Tue 11:30 H 2053

Magnetic measurements on a metal-organic spin-1/2 dimer system – a possible candidate for a 2D field-induced phase transition — •Ulrich Tutsch[1], Bernd Wolf[1], Michael Lang[1], Tonia Kretz[2], Hans-Wolfram Lerner[2], Matthias Wagner[2], Stefan Wessel[3], Tanusri Saha-Dasgupta[4], Harald Jeschke[5], and Roser Valenti[5] — [1]Physikalisches Institut, J.W. Goethe-Universität, SFB/TR49, D-60438 Frankfurt(M), Germany — [2]Institut für Anorganische Chemie, J.W. Goethe-Universität, SFB/TR49, D-60438 Frankfurt(M), Germany — [3]Institut für Theoretische Physik III, Universität Stuttgart, D-70550 Stuttgart, Germany — [4]S.N. Bose National Centre for Basic Sciences, Salt Lake City, Kolkata 700098, India — [5]Institut für Theoretische Physik, J.W. Goethe-Universität, SFB/TR49, D-60438 Frankfurt(M), Germany

We present new data for the low-temperature (T<1K) susceptibility of $C_{36}H_{48}Cu_2F_6N_8O_{12}S_2$ (TK91) as function of the magnetic field. In this material Cu^{2+} ions form spin-1/2 dimers with an intradimer exchange interaction of $J_1/k_B \approx 10$K mediated by a hydroquinone-derived linker. Previous studies have revealed a weak interdimer coupling $J_2/k_B \sim 1$K. Density Functional Theory calculations support these findings and furthermore indicate a quasi 2-dimensional structure of the interdimer coupling. Therefore, we compare the new susceptibility data with Quantum Monte Carlo Simulations for 2-dimensionally coupled dimers. The results indicate that this material is a candidate for a 2D field-induced phase transition.

TT 13.9 Tue 11:45 H 2053

Magnetic properties of the spin-1/2 chain material (6MAP)$CuCl_3$ — •M. Ozerov[1], E. Čižmár[1], S. Zvyagin[1], C. Landee[2], M. Turnbull[2], and J. Wosnitza[1] — [1]Hochfeld-Magnetlabor Dresden (HLD), Forschungszentrum Dresden - Rossendorf, Germany — [2]Clark University, Worcester, MA, USA

Recently, low-dimensional spin systems have received a considerable amount of attention due to their relevance to numerous quantum phenomena such as quantum criticality problems, spin-Peierls transitions, etc. Here we report on magnetization, electron paramagnetic resonance (EPR) and specific-heat measurements of the spin-1/2 Heisenberg antiferromagnetic chain material (6MAP)$CuCl_3$. Magnetization data measured at 0.1 T exhibit a maximum at about 70 K, indicating the low-dimensional character of the magnetic interactions. The data are in a good agreement with the temperature dependence of the resonance peak intensity measured at 73 GHz. At low temperatures (T < 25 K) the EPR linewidth drastically increases, indicating a possible enhancement of 3D short-range-order correlations. Such behavior is consistent with a broad maximum in the specific heat observed at about 1.5 K, which can be interpreted in terms of 3D magnetic ordering. In addition, we present results of room-temperature X-band EPR measurements of (6MAP)$CuCl_3$, including angular dependence of the g-factor and of the resonance linewidth.

The work was supported in part by the DFG through Grant No. ZV 6/1-1.

TT 13.10 Tue 12:00 H 2053

Magnetic properties of the Haldane-gap material NENB — •Erik Čižmár[1], Mikhaylo Ozerov[1], Oleg Ignatchik[1], Thomas P. Papageorgiou[1], J. Wosnitza[1], S. A. Zvyagin[1], Jurek Krzystek[2], Zhixian Zhou[3], Christopher P. Landee[4], Brian R. Landry[4], Mark M. Turnbull[4], and Jan L. Wikaira[5] — [1]Hochfeld-Magnetlabor Dresden (HLD), Forschungszentrum Dresden-Rossendorf, 01314 Dresden, Germany — [2]National High Magnetic Field Laboratory, Tallahassee, USA — [3]Department of Physics and Astronomy, Wayne State University, Detroit, USA — [4]Department of Physics and Carlson School of Chemistry, Clark University, Worcester, USA — [5]Department of Chemistry, University of Canterbury, Christchurch, New Zealand

Results of magnetization and high-field ESR studies of the new spin-1 Haldane-chain material $[Ni(C_2H_8N_2)_2NO_2](BF_4)$ (NENB) are reported. A definite signature of the Haldane state in NENB was obtained. From the analysis of the frequency-field dependence of magnetic excitations in NENB, the spin-Hamiltonian parameters were calculated, yielding $\Delta/k_B = 17.4$ K, $g_\parallel = 2.14$, $D/k_B = 7.5$ K, and $|E/k_B| = 0.7$ K for the Haldane gap, g factor. and the crystal-field anisotropy, respectively. The presence of fractional $S = 1/2$ chain-end states, revealed by ESR and magnetization measurements, is found to be responsible for spin-glass-freezing effects. In addition, extra states in the excitation spectrum of NENB have been observed in the vicinity of the Haldane gap, whose origin is discussed.

The work was supported in part by the DFG through Grant No. ZV 6/1-1.

TT 13.11 Tue 12:15 H 2053

New natural spin-1/2 kagomé systems — kapellasite $Cu_3Zn(OH)_6Cl_2$ and haydeeite $Cu_3Mg(OH)_6Cl_2$ — •Oleg Janson and Helge Rosner — Max Planck Institute for Chemical Physics of Solids, Nöthnitzer str. 40, 01187 Dresden

New natural spin-1/2 systems with kagomé layers — kapellasite $Cu_3Zn(OH)_6Cl_2$ and haydeeite $Cu_3Mg(OH)_6Cl_2$ — are studied by full potential density functional calculations using the fplo6.00-24 code. The band structure, obtained by a paramagnetic calculation, was used to solve a tight-binding model. The transfer integrals were mapped subsequently to a Hubbard model and to a Heisenberg model, giving an estimate for the antiferromagnetic (AF) exchange. The total exchange, containing AF and ferromagnetic (FM) parts, was derived from LSDA + U supercell calculations. As the main result, we find that in both compounds only two exchange integrals are relevant: the nearest neighbour exchange J_1 and the interaction J_d along the diagonals of the Cu^{2+} hexagons. Surprisingly, the size of these integrals depends strongly on the O—H bond length which was therefor optimized with respect to the total energy, resulting in about 1 Å for both compounds. Using the optimized O—H bond length, we find $J_1 > J_d$ in kapellasite and $J_1 \sim J_d$ in haydeeite. According to our results, kapellasite can be described as a modified kagomé lattice, while interpenetrating chains should be considered for haydeeite. Our results should encourage new experimental studies of these interesting materials.

TT 13.12 Tue 12:30 H 2053

Charge excitations and local magnetism: Li_2CuO_2 — •Stefan-Ludwig Drechsler[1], Jiří Málek[1,2], Martin Knupfer[1], Ulrike Nitzsche[1], Helge Rosner[3], and Helmut Eschrig[1] — [1]IFW-Dresden, D-01171 Dresden — [2]Institute of Physics, ASCR, Prague — [3]Max-Planck-Inst. f. Chem. Phys. Fester Stoffe

Electron energy loss spectroscopy and optical conductivity data of the frustrated edge-shared chain cuprate Li_2CuO_2 are reanalyzed within exact diagonalizations of multiband Hubbard models for CuO_2 chains taking into account Cu $3d_{xy}$, $4s$ and/or $4p_{x,y}$ and O $2p_{x,y}$ orbitals. We show that Zhang-Rice (ZR) singlet charge excitations which are generic

for most cuprates are strongly suppressed in favor of ZR triplets as a consequence of substantial ferromagnetic correlations at T=0. A significant suppression also at T=300 K is obtained approximately. The frequency region above 6eV is dominated by O $2p$ to Cu $4p_y$, $4s$ and $4p_x$ transitions. The cluster mapping of these multiband Hubbard models on the Heisenberg J_1-J_2 model is in accord with a total energy analysis of various magnetic structures within LSDA+U FPLO calculations. According to all these theoretical results the observed ferromagnetic ordering of Li_2CuO_2 is caused already by a strong nearest-neighbor (CuO_4 plaquette) ferromagnetic in-chain interaction $J_1 \approx$ -200 K compared with the next-nearest neighbor exchange $J_2 \approx 35$ K and it is only a bit further stabilized by a specific frustrating weak antiferromagnetic interchain exchange in contrast with previous studies [1].

[1] Y. Mizuno *et al.*, Phys. Rev. B **57**, 5326 (1998).

TT 13.13 Tue 12:45 H 2053

Competing exchange interactions in the one-dimensional spin-1/2 system Li_2CuO_2 — •Ulrike Nitzsche[1], Stefan-Ludwig Drechsler[1], and Helge Rosner[2] — [1]IFW Dresden, P.O. Box 270116, D-01171 Dresden — [2]MPI CPfS Dresden, Nöthnitzer Straße 40, D-01187 Dresden

The spin 1/2 chain-cuprate Li_2CuO_2 is the archetype for competing nearest-neighbor and next-nearest-neighbor exchange interaction J_1 and J_2 along the CuO_2 chains. The magnitude of J_1 and J_2, especially their ratio α is crucial for the understanding of the magnetic ground state and is controversially disputed since a decade. Here, we report a density functional based electronic structure study to evaluate the leading in-chain and inter-chain exchange constants. We combine an LDA+U total energy approach for different spin configurations with the results from a tight-binding fit mapped onto a Heisenberg model. Our results yield a ferromagnetic ground state with a ferromagnetic J_1 and an antiferromagnetic J_2 and an α value well inside the region of the ferromagnetically ordered phase. A brief comparison with results from literature is given.