TT 18: Correlated Electrons: Low-dimensional Systems - Materials 2

Time: Tuesday 15:15–18:45 Location: H 2053

TT 18.1 Tue 15:15 H 2053

Charge Ordering in $(TMTTF)_2X$: Evidences for Structural Changes — •Mariano de Souza[1], Alec Moradpour[2], Pascale Foury[2], Jean-Paul Pouget[2], and Michael Lang[1] — [1]Physikalisches Institut, J.W. Goethe-Universität, Max-von-Laue Str. 1, SFB/TRR49, D-60438 Frankfurt am Main, Germany — [2]Laboratoire de Physique des Solides, Université Paris Sud, CNRS, UMR 8502, 91405 Orsay, France

The $(TMTTF)_2X$ family of quasi-1D organic conductors has been the subject of intensive studies over the last two decades. Recently, NMR spectroscopy studies [1] revealed the existence of a charge-ordered (CO) phase in the X=PF_6 and AsP_6 salts at $T_{CO}\approx$65 and 105 K, respectively. Until now, however, no evidence of structural changes has been reported in the literature and, due to this, the latter has been referred to as a "*structureless*" phase transition. In this contribution, we present directional-dependent high-resolution thermal expansion measurements on the X=PF_6 salt. Our findings reveal strong anisotropic lattice effects at both the Spin-Peierls and the CO transition, with the strongest response along the c^*-axis, along which the stacks are separated by the PF_6^- anions and where the transfer integral t_{c^*} is very weak. For the chain direction (a-axis), less pronounced signatures are observed. These results provide the first evidence for structural effects at the CO transition, indicating both charge and lattice degrees of freedom are involved in this transition.

[1] D.S. Chow *et al.*, Phys. Rev. Lett. **85**, 1698 (2000).

TT 18.2 Tue 15:30 H 2053

Realistic Parameters for the description of the two-dimensional molecular metal Θ-BEDT-TTF — •Andreas Dolfen[1], Erik Koch[1], Laura Cano-Cortes[2], and Jaime Merino[2] — [1]Institut für Festkörperforschung, Forschungszentrum Jülich, Germany — [2]Departamento de Física Teórica de la Materia Condensada,

In order to study correlations in the two dimensional molecular metal Θ-$(BEDT\text{-}TTF)_2I_3$ we employ density-functional theory to calculate realistic parameters for extended Hubbard models.

We evaluate the hopping matrix elements for different angles and distances of two molecules observing a surprising sensitivity with respect to certain molecular orientations. This is because a large part of the molecular orbital is situated next to the sulphur atoms outside the molecules.

The Coulomb parameters, however, are less sensitive. We calculate the on-site as well as the longer range Coulomb repulsion including intra-molecular screening and screening due to all the other molecules (BEDT-TTF and I_3).

TT 18.3 Tue 15:45 H 2053

First principles study of the charge transfer salt κ-$(BEDT\text{-}TTF)_2Cu(CN)_3$ — •Harald O. Jeschke, Hem C. Kandpal, and Roser Valenti — Institut für Theoretische Physik, Universität Frankfurt, Max-von-Laue-Str. 1, 60438 Frankfurt, Germany

The charge transfer salt κ-$(BEDT\text{-}TTF)_2Cu(CN)_3$ has attracted a lot of attention due to experimental evidence that it is a realization of a spin liquid: no magnetic ordering was found down to 20 mK [1]. For a good description of this behaviour, it is crucial to consider the appropriate effective model for this system. Here, we present electronic structure calculations in the frame of density functional theory (DFT) and derive an effective model with the NMTO (N-th order muffin tin orbital) downfolding method and discuss its features. Since from X-ray diffraction, the structure of κ-$(BEDT\text{-}TTF)_2Cu(CN)_3$ has been determined without hydrogen positions, we first prepare a very similar structure including hydrogen atoms and carefully relax it using the projector augmented wave method. This structure is then used for the DFT and NMTO analysis.

[1] S. Ohira, Y. Shimizu, K. Kanoda and G. Saito, J. Low Temp. Phys. **142**, 153 (2006).

TT 18.4 Tue 16:00 H 2053

Extending Grüneisen's relation on systems with two magnetic energy scales — •Andreas Brühl[1], Bernd Wolf[1], Christoph Gross[1], Wolf Assmus[1], Roser Valenti[2], Stefan Glocke[3], Andreas Klümper[3], and Michael Lang[1] — [1]Physikalisches Institut, Universität Frankfurt, D-60438 Frankfurt(M) — [2]Institut für Theoretische Physik, Universität Frankfurt, D-60438 Frankfurt(M) — [3]Theoretische Physik, Universität Wuppertal, D-42097 Wuppertal

The high-pressure phase of $(VO)_2P_2O_7$ (abbr. HP-VOPO) consists of alternating S=1/2 spin chains with exchange constants J_1 and J_2 of similar size (≈ 120 K). This gives rise to a second, well-separated energy scale in the system, the spin gap $\Delta = 32$ K. Thermal expansion measurements on HP-VOPO reveal distinct anomalies related to these two energy scales. However, the relative size of the low-temperature anomaly, corresponding to the spin gap, is very much enlarged compared to its size in the specific heat. Therefore, the commonly used Grüneisen relation, assuming a proportionality between the magnetic contributions α_{mag} and C_{mag} to thermal expansion and specific heat, does not apply. Instead, an extended form of this relation is presented, which contains an additional term in α_{mag} proportional to the derivative of the magnetic entropy with respect to the spin gap. Using accurate T-DMRG calculations of the required thermodynamic quantities, this leads to a good description of the thermal expansion of HP-VOPO in the whole temperature range. This method could be generalized to cover other spin systems with two or more exchange constants, e. g. to determine their values via thermal expansion measurements.

TT 18.5 Tue 16:15 H 2053

Electron-phonon interaction in the lamellar cobaltate Na_xCoO_2 — •Alexander Donkov[1], Maxim Korshunov[1,2], and Ilya Eremin[1,3] — [1]Max-Planck-Institut für Physik komplexer Systeme, D-01187 Dresden, Germany — [2]L.V. Kirensky Institute of Physics, Siberian Branch of Russian Academy of Sciences, 660036 Krasnoyarsk, Russia — [3]Institute für Mathematische und Theoretische Physik, TU Braunschweig, D-38106 Braunschweig, Germany

We study theoretically the dependence of the electron-phonon interaction in Na_xCoO_2 on the sodium concentration, x. For the two phonon modes found in Raman experiments, A_{1g} and E_{1g}, we calculate the matrix elements of the electron-phonon interaction. Analyzing the feedback effect of the conduction electrons on the phonon frequency, ω, we compare the calculated and experimentally observed doping dependence of the A_{1g} mode. Furthermore, due to the momentum dependence of the electron-phonon coupling for the E_{1g} symmetry we find no renormalization of the corresponding phonon frequency which agrees with experiment. Our results shed light on the possible importance of the electron-phonon interaction in this system.

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TT 18.6 Tue 16:30 H 2053

Effect of composition on phonon anomalies in $Na_xCoO_2 \cdot yH_2O$ — •Peter Lemmens[1,2], Dietrich Wulferding[1], Patric Scheib[1], Kwang-Yong Choi[3], Vladimir Gnezdilov[4], Fangcheng Chou[5], Chengtian Lin[2], and Bernhard Keimer[2] — [1]IPKM, TU Braunschweig — [2]MPI-FKF, Stuttgart — [3]NHMFL, Florida — [4]B.I. Verkin Inst. for Low Temp. Phys., NASU, Kharkov — [5]CMSE, MIT, Cambridge, USA

A quantitative analysis of phonon frequencies for nonsuperconducting and superconducting cobaltates $Na_xCoO_2 \cdot yH_2O$ shows pronounced phonon anomalies in the proximity of the charge ordering instabilities in $Na_{0.5}CoO_2$. Furthermore, as function of Na doping a systematic shift of a high energy out-of-plane oxygen phonon is observed in a wide doping range. The corresponding in-plane mode does not show any appreciable anomaly.

Work supported by DFG, ESF-HFM and MRSEC Program of NSF under award number DMR 02-13282.

TT 18.7 Tue 16:45 H 2053

On the metallic conductivity of the delafossites $PdCoO_2$ and $PtCoO_2$ — •Volker Eyert[1,2], Raymond Frésard[1], and Antoine Maignan[1] — [1]Laboratoire CRISMAT, UMR CNRS-ENSICAEN(ISMRA) 6508, 6 Boulevard Maréchal Juin, 14050 Caen Cedex, France — [2]Center for Electronic Correlations and Magnetism, Institut für Physik, Universität Augsburg, 86135 Augsburg, Germany

The origin of the quasi two-dimensional behavior of $PdCoO_2$ and $PtCoO_2$ is investigated by means of electronic structure calculations. They are performed using density functional theory in the local density approximation as well as the new full-potential augmented spherical

wave method. We show that the electric conductivity is carried almost exclusively by the in-plane Pd (Pt) *d* orbitals. In contrast, the insulating O–Co–O sandwich layers of octahedrally coordinated Co atoms may be regarded as charge carrier reservoirs. This leads to a weak coupling of the Pd (Pt) layers and causes the strong anisotropy of the electric conductivity.

15 min. break

TT 18.8 Tue 17:15 H 2053

Thermal conductivity of TiOX (X=Br,Cl) — •Nikolai Hlubek[1], Christian Hess[1], Bernd Büchner[1], Michael Sing[2], Ralph Claessen[2], Sander van Smaalen[3], and Paul van Loosdrecht[4] — [1]IFW Dresden, Germany — [2]Experimentelle Physik 4, Universität Würzburg, Germany — [3]Laboratory of Crystallography, Universität Bayreuth, Germany — [4]Zernike Institute for Advanced Materials, University of Groningen, Netherlands

We report experimental results on the thermal conductivity $\kappa(T)$ of the compounds TiOBr and TiOCl. Below room temperature the compounds undergo two phase transitions T_{c2} and T_{c1}. Above T_{c2} the compounds should contain S=1/2 spin-chains with $J_{Cl} = 676K$ and $J_{Br} = 375K$ respectively formed by direct orbital overlap of the Ti-atoms. Below T_{c1} the chains dimerise to form a non-magnetic ground state. Our data exhibits pronounced anomalies at T_{c2} and T_{c1} confirming the transitions being of second and first order respectively. Surprisingly, $\kappa(T)$ appears to be dominated by phonon heat conduction, since we don't find indications of significant magnetic contributions. This is in contrast to the expectation of a spin chain system. In this context we discuss possible scenarios to understand the unusual behaviour of the thermal conductivity.

TT 18.9 Tue 17:30 H 2053

Competing exchange interactions in the edge-shared chain cuprates $CuCl_2$ and $CuBr_2$ — •Miriam Schmitt[1], Stefan-Ludwig Drechsler[2], and Helge Rosner[1] — [1]MPI CPfS Dresden, Nöthnitzer Strasse 40, D-01187 Dresden — [2]IFW Dresden, P.O.Box 270116, D-01171 Dresden

Low dimensional spin 1/2 chain systems show fascinating phase diagrams with many unusual magnetic ground states as spin-Peierls, spin gap or helically ordered states. These ground states are usually driven by the strong competition between the nearest and next nearest neighbor in-chain interactions. We will present a detailed study of the electronic and magnetic properties based on density functional calculation for the edge shared chain Cu^{2+} compounds $CuCl_2$ and $CuBr_2$. Starting from LDA band structure calculation we developed an effective one-band tight-binding model and mapped it subsequently to a Heisenberg model to evaluate the orbitals and main interactions relevant for the low energy properties. The combination of these results with the exchange integrals from LDA+U total energy differences of different spin configurations leads to reliable microscopic models. For $CuCl_2$ and $CuBr_2$ we find a strong frustration due to competing antiferromagnetic J_2 and ferromagnetic J_1. Unless the significant interchain coupling stabilizes a commensurate ground state we predict that both compounds exhibit a helically ordered state at low temperatures.

TT 18.10 Tue 17:45 H 2053

Electric field gradients in low dimensional cuprates - a comparative study — •Katrin Koch[1], Klaus Koepernik[1,2], and Helge Rosner[1] — [1]MPI-CPFS, Dresden — [2]IFW, Dresden

The theoretical description of strongly correlated systems is still a challenging task. A possible approach to this problem resulting in realistic electron densities is the LSDA+U method where the strong Coulomb repulsion U is treated in a mean field like approximation. Unfortunately the parameter U is not known and needs to be evaluated, i.e. in comparison of calculated properties and experimental measurements.

Here, we present calculations for the electric field gradients (V_{zz}) on the Cu^{2+} sites in strongly correlated low dimensional cuprates. By comparing the calculated V_{zz}(U) with the V_{zz} from NMR experiments for these compounds we can evaluate U. To avoid numerical ambiguity we used two different DFT-band structure schemes: FPLO and WIEN2k. The results are consistent with respect to each other: In part we find good agreement with resulting values of U from other evaluation procedures, especially for strongly distorted Cu^{2+} environment (e.g. $Cu_2P_2O_6CH_2$), though considerable deviation in direction of smaller U values for other compounds (e.g. La_2CuO_4) is obtained. Possible causes for this deviation will be discussed.

TT 18.11 Tue 18:00 H 2053

Temperature dependence of a zero bias anomaly in scanning tunnelling spectra of $Sr_4Ru_3O_{10}$ — •Bernhard Nansseu[1], Tatjana Novgorodov[1], Michael Waelsch[1], Jürgen Hager[1], Jiandi Zhang[2], R. Moore[3], Ward Plummer[3], Zhiqiang Mao[4], and Rene Matzdorf[1] — [1]Universität Kassel, Kassel, Germany — [2]Florida International University, Miami, USA — [3]Oak Ridge National Laboratory and University of Tennessee, USA — [4]Tulane University, New Orleans, USA

We have studied a zero bias anomaly in scanning tunnelling spectra of the layered ruthenate $Sr_4Ru_3O_{10}$. This material shows a dip-like feature in the dI/dV spectra, which has previously observed in the single-layer Sr_2RuO_4 and double-layer $Sr_3Ru_2O_7$ ruthenates. We have studied in particular the temperature dependence of the zero bias anomaly, which is in all three materials different. The triple-layer material shows intergrowth of single and double layers, which have been identified by their spectroscopic fingerprint. Finally, we discuss different effects as possible explanations for the zero bias anomaly.

TT 18.12 Tue 18:15 H 2053

Temperature dependence of the anomalous exponent in $Li_{0.9}Mo_6O_{17}$that reveals Luttinger Liquid behavior — Tatjana Novgorodov[1], Bernard Nansseu[1], Michael Waelsch[1], Jian He[2], Rongying Jin[2], David Mandrus[2,3], and •Rene Matzdorf[1] — [1]Universität Kassel, Kassel, Germany — [2]Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA — [3]The University of Tennessee, Knoxville, Tennessee, USA

Scanning tunnelling spectroscopy (STS) has been used to study the Luttinger-liquid behavior of the purple bronze $Li_{0.9}Mo_6O_{17}$ in the temperature range 5K¡T¡300K. In the entire temperature range the suppression of density of states at the Fermi-energy could be fitted very good by a model describing the tunneling into a Luttinger liquid at ambient temperature. The power-law exponent extracted from these fits reveals a significant increase above 200K. It changes from alpha=0.6 at low temperature to alpha=1.0 at room temperature.

TT 18.13 Tue 18:30 H 2053

Microscopic simulation and analysis of a spin crossover transition — •Harald O. Jeschke[1], L. Andrea Salguero[1], Badiur Rahaman[2], Christian Buchsbaum[3], Volodymyr Pashchenko[3], Martin U. Schmidt[3], Tanusri Saha-Dasgupta[2], and Roser Valenti[1] — [1]Institut für Theoretische Physik, Universität Frankfurt, Max-von-Laue-Str. 1, 60438 Frankfurt, Germany — [2]S.N. Bose National Centre for Basic Sciences, JD Block, Sector 3, Salt Lake City, Kolkata 700098, India — [3]Institut für Anorganische und Analytische Chemie, Universität Frankfurt, Max-von-Laue-Str. 7, 60438 Frankfurt, Germany

In spin crossover materials, an abrupt phase transition between a low spin state and a high spin state can be driven by temperature, pressure or illumination. Of a special relevance are Fe(II) based coordination polymers where, in contrast to molecular systems, the phase transition shows a pronounced hysteresis which is desirable for technical applications. A satisfactory microscopic explanation of this large cooperative phenomenon has been sought for a long time. The lack of X-ray data has been one of the reasons for the absence of microscopic studies. In this work, we present an efficient route to prepare reliable model structures and within *ab initio* density functional theory analysis and effective model considerations we show that in polymeric spin crossover compounds magnetic exchange between high spin Fe(II) centres is as important as elastic couplings for explaining the considerable cooperativity and thus the width of the hysteresis.