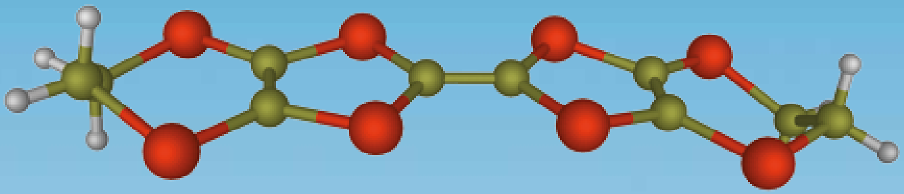

ISCOM 2015

11TH INTERNATIONAL SYMPOSIUM

ON CRYSTALLINE ORGANIC METALS, SUPERCONDUCTORS AND MAGNETS

SEPTEMBER 6 – 11, 2015 BAD GÖGGING, GERMANY

CONFERENCE PROGRAM

CONTENTS

Cover: Personenschifffahrt Stadler, Kelheim

WELCOME MESSAGE

Dear ISCOM 2015 participants,

On behalf of the organizing committee, we welcome you in Bad Gögging to the 2015 International Symposium of Crystalline Organic Metals, Superconductors and Magnets (ISCOM 2015). The symposium will provide an interdisciplinary forum for discussing the most recent developments in the physics, chemistry, material science technology and modeling of molecular solids.

We are very honored to host this 11th event of an ISCOM symposium which continues a long-lasting series of meetings initiated in Mittelberg (1995), and followed by Sesimbra (1997), Oxford (1999), Rusutsu (2001), Port-Bourgenay (2003), Key West (2005), Peniscola (2007), Niseko (2009), Poznan-Gniezno (2011) and Montreal (2013).

The ISCOM 2015 will be held in Bad Gögging in Bavaria close to the famous Danube Gorge. It will be the occasion for chemists and physicists of all over the world to present their latest contributions to the lively field of synthesis and physics of crystalline organic molecular solids.

In this Abstract Booklet you will find the conference program and the abstracts of all contributions that will be presented during the symposium. On behalf of the organizers we would like to thank the members of the International Advisory Committee for their suggestions and support.

We wish you a fruitful and pleasant stay in Bad Gögging.

Michael Lang
Chair ISCOM 2015

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[*] *James Brooks passed away in September 2014. The organic community is mourning for an excellent scientist with an engaging personality.*

ACKNOWLEDGEMENT

We want to thank the following sponsors for their contribution to the success of the symposium:

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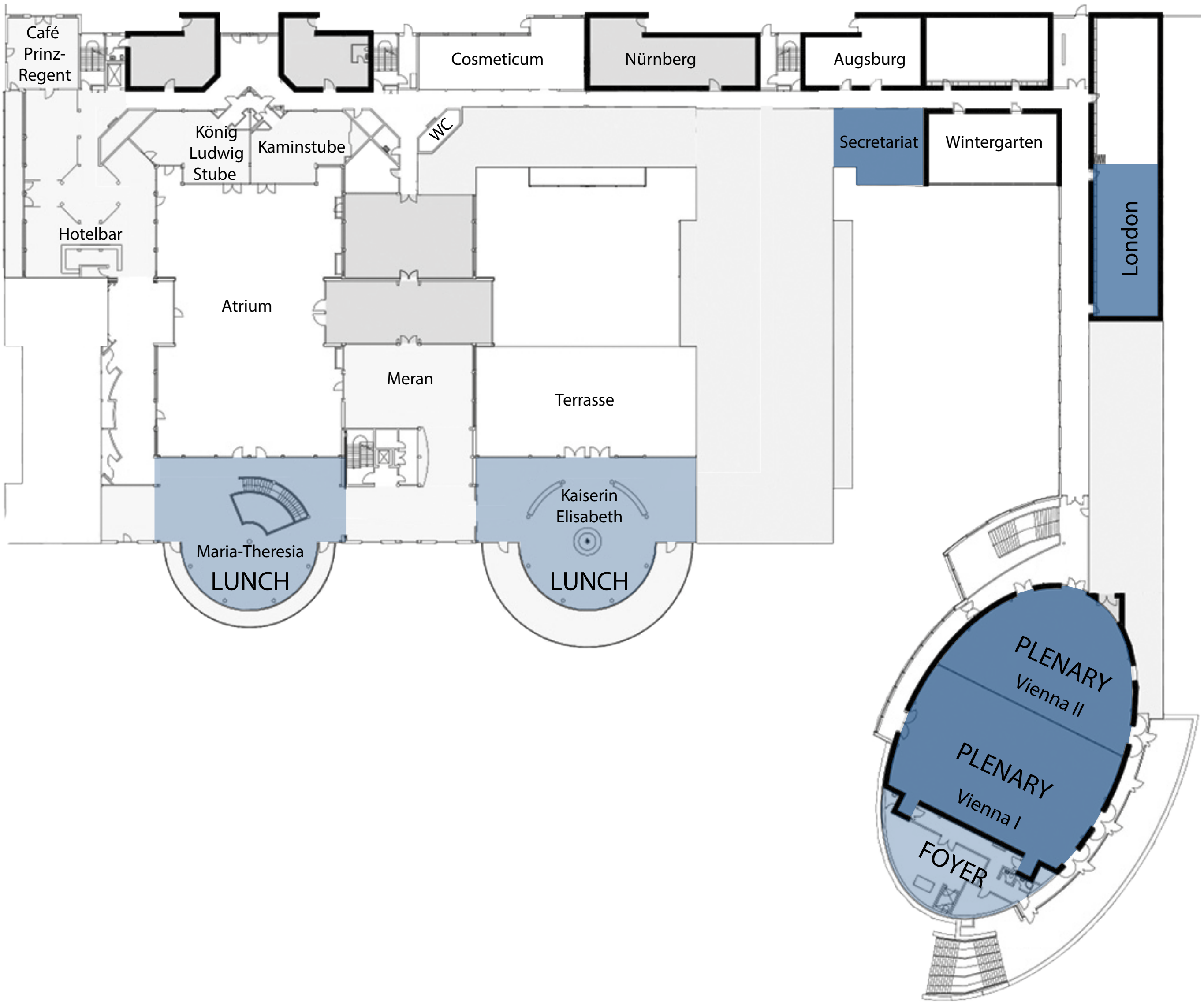
HOTEL FLOOR MAP



INFORMATION

REGISTRATION

The registration desk is located in the Hotel lobby close to the reception.

Each ISCOM participant will receive a symposium kit upon registration.
The registration fee comprises

- Admissions to all sessions, poster periods and program.
- All the meals, refreshment breaks and welcome reception at the hotel.
- Admission to the excursion and banquet event.

MEALS

Breakfast, lunch and dinner will be served in the main restaurant located on the first floor.

NAME BADGES

Participants and accompanying persons are required to wear the official symposium name badge on all symposium occasions.

REFRESHMENTS

Refreshments are served in the Foyer Vienna during breaks scheduled between the oral sessions in the morning and afternoon.

INTERNET ACCESS

Wireless is available throughout the conference room Vienna and its foyer. The access to the internet in all rooms of the hotel will be charged extra with 7,-- € per day.

SECRETARIAT

The secretariat office will be located in room No. 2795 on the first floor opposite to the conference room “Augsburg”.
Opening hours during coffee breaks and lunch time (12:30h – 13:30h).

DISCLAIMER

The organizers are not liable for damages and/or losses of any kind which may be incurred by the Conference delegates or by any other individuals accompanying them, both during the official activities as well as going to/from the Symposium. Delegates are responsible for their own safety and belongings.

INSTRUCTIONS FOR SPEAKERS

TIME ALLOCATION FOR ORAL TALKS

Invited:	30 min = 25 min + 5 min (discussion)
Contributed:	15 min = 12 min + 3 min (discussion)

Oral presentations can be done by using your own PC or MAC computers. Mac owners should not forget to bring their display connector. You can also upload your (Power Point or pdf format) presentation on the PC available in the session room. Speakers are asked to hand over their laptop to the present technician in the backside of the auditorium or upload their presentation during the break that precedes their session. Please note that due to technical reasons and for best results on the screen the format of your presentation should be in 4 x 3. Speakers are requested to sit in the first row of the session room close to the stage at least 20 minutes before their presentation.

INSTRUCTIONS FOR POSTER PRESENTATIONS

The size of each poster on boards must not exceed 115 cm (width) x 145 cm (height). A Poster number is assigned to each presentation. Pushpins will be provided on site.

SCHEDULE

Poster Session I
Tuesday, September 8
Afternoon: Poster Installation
Session: 20:00 – 22:00 (Vienna Foyer)
Wednesday Morning, September 9
Poster Removal

Poster Session II
Thursday, September 10
Afternoon: Poster Installation
Session: 20:00 – 22:00 (Vienna Foyer)
Friday Morning, September 11
Poster Removal

SUNDAY, SEPTEMBER 6, 2015

15:00 - 19:00	ISCOM Check-in
19:00 - 21:00	Dinner + Welcome Reception

MONDAY, SEPTEMBER 7, 2015

Oral Sessions

Location: Vienna I + II

08:50 - 09:00	Opening	Michael Lang
Session 1	**Superconductivity I**	
09:00 - 09:30	Molecular layers in iron-based superconductors	S. J. Blundell *University of Oxford, Great Britain*
09:30 - 10:00	Magnetism and electron-phonon interaction in the emergence of organic superconductivity	C. Bourbonnais *Université de Sherbrooke, Canada*
10:00 - 10:15	Disorder-induced gap in the normal density of states of the organic superconductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br	H. - J. Elmers *Universität Mainz, Germany*
10:15 - 10:30	Electron correlation induced superconductivity in the quarter-filled band	*R. T. Clay* *Mississippi State University, USA*
10:30 - 11:00	Coffee Break	

Session 2	**Design of New Materials I**	
11:00 - 11:30	Controlling crystallographic and electronic structure in cationic and anionic radical salts	J. A. Schlueter *National Science Foundation, USA*
11:30 - 12:00	Intramolecular charge transport through organic radicals	J. Veciana *Institut Ciencia Materials Barcelona, Spain*
12:00 - 12:30	Structural and magnetic properties of thiazyl coordination complexes	K. Preuss *University of Guelph Chemistry, Canada*
12:30 - 14:00	Lunch	

Session 3	**Functional Materials**	
14:00 - 14:30	Intrinsic electrostatic control of TTF-TCNQ molecular layers on noble metal surfaces	P. Maksymovych *Oak Ridge National Laboratory Center for Nanophase Materials Sciences, USA*
14:30 - 15:00	Anilate-based functional molecular materials with conducting and magnetic properties	M. L. Mercuri *University of Cagliari, Italy*
15:00 - 15:30	Organic phase-transition transistors with strongly-correlated electrons	H. Yamamoto *Institute for Molecular Science Research Center of Integrative Molecular Systems, Japan*
15:30 - 15:45	Conductors and transistors based on BTBT-based charge-transfer complexes	T. Mori *Tokyo Institute of Technology Japan*
15:45 - 16:00	Preparation of ultrathin films of electrically active Metal-Organic Frameworks	V. Rubio-Giménez *Universitat de València, Spain*
16:00 - 16:15	Intramolecular electronic transport studied with organic radicals	C. Rovira *Institut de Ciència de Materials de Barcelona (ICMAB-CSIC)/CIBER-BBN Nanomol, Spain*
16:15 - 16:45	Coffee Break	

Session 4	**Mott Physics I**	
16:45 - 17:15	Diverse manifestation of electron correlation in molecular materials	K. Kanoda *University of Tokyo, Japan*
17:15 - 17:30	Pseudogap phase and the Mott transition in organic and cuprate superconductors	J. Merino *Universidad Autonoma de Madrid, Spain*
17:30 - 17:45	Length changes, critical properties and role of electron-phonon coupling at the Mott transition in κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl	E. Gati *Goethe Universität, Germany*
17:45 - 18:00	Exploring Mott criticality in κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br by utilizing glasslike structural ordering	B. Hartmann *Goethe Universität, Germany*
19:00 - 20:00	Dinner	
20:00 - 21:30	Advisory Board Meeting - Room London	

TUESDAY, SEPTEMBER 8, 2015

Oral Sessions

Location: Vienna I + II

Session 5	**Dirac Electron Systems**	
09:00 - 09:30	Single-component molecular conductor with Dirac cones	R. Kato *RIKEN Condensed Molecular Materials Laboratory, Japan*
09:30 - 10:00	Spin-Order in molecular massless dirac fermion systems	N. Tajima *Toho University Physics, Japan*
10:00 - 10:15	Experimental determination of anisotropy of dirac cone and van Hove singularity in the organic dirac fermion system	T. Osada *University of Tokyo, Japan*
10:15 - 10:30	Designing molecular crystals using symmetry and emergence: A study in $Mo_3S_7(dmit)_3$	A. Jacko *The University of Queensland School of Mathematics and Physics, Australia*
10:30 - 11:00	Coffee Break	

Session 6	**Charge Order I**	
11:00 - 11:30	Charge glass in θ-$(ET)_2X$ - Emergent phenomena and functionalities	F. Kagawa *RIKEN Center for Emergent Matter Science (CEMS), Japan*
11:30 - 12:00	Phase diagram of the triangular extended Hubbard model	L. F. Tocchio *International School for Advanced Studies (SISSA), Italy*
12:00 - 12:15	Tilted stripe-type charge order and destabilization of the threefold state in θ-$(BEDT\text{-}TTF)_2X$	H. Seo *RIKEN Condensed Matter Theory Laboratory, Japan*
12:15 - 12:30	Optical investigation of charge fluctuations in the organic superconductor β''-$(BEDT\text{-}TTF)_4[(H_3O)Ga(C_2O_4)_3]C_6H_5NO_2$	A. Pustogow *Universität Stuttgart, Germany*
12:30 - 14:00	Lunch	

Session 7	**Quantum Spin Liquids I**	
14:00 - 14:30	Insight from computational approaches into superconductors and spin liquids in molecular solids	M. Imada *University of Tokyo, Japan*
14:30 - 14:45	New quantum spin-liquid candidates, valence-bond solid and superconductors κ-$(BEDT\text{-}TTF)_2X$ ($X=Ag_2(CN)_3$, $B(CN)_4$, CF_3SO_3, $Ag(CN)[N(CN)_2]$, etc.)	G. Saito *Meijo University, Japan*
14:45 - 15:00	Quantum spin systems in κ-ET salts	M. Maesato *Kyoto University, Japan*
15:00 - 15:15	Propagating spin excitations in low-dimensional spin liquids investigated using µSR	F. Pratt *STFC ISIS Muon Group, Great Britain*
15:15 - 15:30	Ambient pressure superconductivity in nanopatterned crystals of quantum spin liquid κ-$(ET)_2Cu_2(CN)_3$	A. A. Bardin *Russian Academy of Sciences - Chernogolovka, Russia*
15:30 - 15:45	Dual-functional molecular crystal from candidate quantum spin liquid with Jahn-Teller distortion	B. Zhang *Chinese Academy of Sciences, China*
15:45 - 16:15	Coffee Break	

Session 8	**Magnetic Field Effects**	
16:15 - 16:45	Hall resistance anomalies in two dimensional organic conductor α-$(BEDT\text{-}TTF)_2KHg(SCN)_4$	S. Uji *National Institute for Materials Science, Japan*
16:45 - 17:00	Interlayer charge transport and dimensional crossovers in a layered organic conductor	M. Kartsovnik *Walther-Meißner-Institut, Germany*
17:00 - 17:15	Spin–charge coupling in a series of π–**d** molecular conductors $(DIETSe)_2FeBr_{4x}Cl_{4(1-x)}$	G. Kawaguchi *Kyoto University, Japan*
17:15 - 17:30	Electronic states of the organic superconductor $(TMTSF)_2FSO_3$ studied by the Hall effect measurements	W. Kang *Ewha Womans University Physics, Korea Republic (South)*
17:30 - 17:45	Search for the superconducting gap structure in $\boldsymbol{\kappa}$-$(BEDT\text{-}TTF)_2\mathbf{X}$ by use of thermal-conductivity measurements	E. L. Green *Hochfeld-Magnetlabor Dresden, Germany*
19:00 - 20:00	Dinner	
20:00 - 22:00	Poster Session I - Vienna Foyer *Afternoon: Poster Installation*	

WEDNESDAY, SEPTEMBER 9, 2015

Oral Sessions

Location: Vienna I + II

Session 9	**Charge Order II**	
09:00 - 09:30	Interplay between structural and electronic effects at the charge ordering transitions of BEDT-TTF and TMTTF quarter-filled organic salts	J.-P. Pouget *Université Paris-sud Laboratoire de Physique des Solides, France*
09:30 - 10:00	Charge redistribution in halogen bonded charge-transfer salts of unsymmetrical TTF derivatives studied by IR and Raman spectroscopy	R. Świetlik *Polish Academy of Sciences Institute of Molecular Physics, Poland*
10:00 - 10:15	Donor-anion interactions at the charge localization and charge ordering transitions of (TMTTF)$_2$AsF$_6$ probed by NEXAFS	K. Medjanik *Lund University, Sweden*
10:15 - 10:30	Charge degrees of freedom under high pressure in an organic dimer-Mott insulator β'-(BEDT-TTF)$_2$ICl$_2$	K. Hashimoto *Tohoku University, Japan*
10:30 - 11:00	Coffee Break	

Session 10	**Superconductivity II**	
11:00 - 11:30	Superconductivity in ß”-(ET)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ at low and high magnetic fields	S. Brown *UCLA Department of Physics & Astronomy, USA*
11:30 - 12:00	Charge density wave-metal (superconductivity) coexistence in the TTF(X(dmit)$_2$)$_2$ family (X=Ni,Pd)	C. Pasquier *Laboratoire de Physique des Solides, France*
12:00 - 12:15	Superconductivity beyond the dimer model in single- and multilayered BEDT-TTF based charge transfer salts	H. O. Jeschke *Goethe-Universität Frankfurt, Germany*
12:30- 13:45	Lunch	

Session 11	**Ferroelectricity I**	
13:45 - 14:15	Charge fluctuation and magnetism in dimer Mott insulators	C. Hotta *University of Tokyo, Japan*
14:15 - 14:30	Covalency-induced electric polarization in tetrathiafulvalene-p-chloranil (TTF-CA) studied by means of maximally-localized Wannier orbitals	S. Ishibashi *AIST Nanomaterials Research Institute, Japan*
14:30 - 14:45	Current switching behaviour of hydrogen-bonding ferroelectrics	T. Akutagawa *Tohoku University, Japan*
15:30 - 22:00	Excursion	

THURSDAY, SEPTEMBER 10, 2015

Oral Sessions

Location: Vienna I + II

Session 12	**Quantum Spin Liquids II**	
09:00 - 09:30	Proton-electron coupled functionalities based upon catechol-TTF based molecular materials	H. Mori *The University of Tokyo, Japan*
09:30 - 10:00	Prediction of a spin-liquid ground state in the geometrically frustrated molecular crystal Mo$_3$S$_7$(dmit)$_3$	B. Powell *University of Queensland, Australia*
10:00 - 10:15	Spatial symmetry breaking in κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ investigated by terahertz emission spectroscopy	K. Itoh *Tohoku University, Japan*
10:15 - 10:30	Spin correlation and dynamics of the spin-liquid material EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ revealed by in-plane ESR measurements	Y. Oshima *RIKEN Condensed Molecular Materials Lab., Japan*
10:30 - 11:00	Coffee Break	

Session 13	**Ferroelectricity II**	
11:00 - 11:30	Donor-acceptor type organic ferroelectric crystals and thin films	S. Horiuchi *National Institute of Advanced Industrial Science and Technology, Japan*
11:30 - 11:45	Novel dielectric, magnetic and magnetodielectric effects in dimer-type organic salts	S. Ishihara *Tohoku University, Japan*
11:45 - 12:00	Electrodynamics and first-principles phonon calculations for the spin-liquid system κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$	S. Tomic *Institut za fiziku, Croatia*
12:00 - 12:15	Revisiting the charge-ordered α-(BEDT-TTF)$_2$I$_3$: anisotropy of electronic properties	M. Basletić *Institut za fiziku, Croatia*
12:15 - 12:30	Dielectric spectroscopy on organic charge-transfer salts	P. Lunkenheimer *University of Augsburg, Germany*
12:30 - 14:00	Lunch	

Session 14	**Design of New Materials II**	
14:00 - 14:30	Development of molecular conductors based on tetramethyl-TTP and its related donors	Y. Misaki *Ehime University, Japan*
14:30 - 15:00	Family of Q2D molecular conductors (BEDT-TTF)$_4$(H$_3$O$^+$)[M^{III}(C$_2$O$_4$)$_3$]×Solvent: mono- and bilayered phases, solvent-depending conductivity, phase transitions	L. Zorina *Institute of Solid State Physics RAS, Russia*
15:00 - 15:15	Bilayer molecular metals (CNB-EDT-TTF)$_4$X: A new prototype of 2D Molecular Conductors	M. Almeida *University of Lisbon, Portugal*
15:15 - 15:30	Chiral conductors based on methylated TTF derivatives	N. Avarvari *CNRS-University, France*
15:30 - 15:45	Molecular alloys of neutral gold/nickel dithiolene complexes in single component semiconductors	M. Fourmigué *Université Rennes 1 & CNRS, ISCR - Rennes, France*
15:45 - 16:15	Coffee Break	

Session 15	**Mott Physics II**	
16:15 - 16:45	Randomness effect on the correlated electrons in the molecular conductors irradiated by X-ray	T. Sasaki *Tohoku University, Japan*
16:45 - 17:00	Detailed study of the Mott transition in EtMe$_3$P[Pd(dmit)$_2$]	M. Abdel Jawad *RIKEN Condensed Molecular Materials Laboratory, Japan*
17:00 - 17:15	Antiferromagnetic fluctuations in organic superconductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br detected by Raman spectroscopy.	N. Drichko *Johns Hopkins University, USA*
17:15 - 17:30	Gold dithiolene complexes as Mott Insulators	D. Lorcy *Université de Rennes, France*
19:00 - 20:00	Dinner	
20:00 - 22:00	Poster Session II - Vienna Foyer *Afternoon: Poster Installation*	

FRIDAY, SEPTEMBER 11, 2015

Oral Sessions

Location: Vienna I + II

SESSION 16	**ULTRAFAST PHENOMENA**	
09:00 - 09:30	Strong field effects on organic conductors induced by nearly single-cycle light pulse	S. Iwai *Tohoku University, Japan*
09:30 - 10:00	Control of electronic interactions in organic conductors and cuperconductors	S. Kaiser *Max Planck Institut Hamburg, Germany*
10:00 - 10:15	Coherent excitations at the neutral-ionic transition: Femtosecond dynamics on diabatic potential energy surfaces	A. Painelli *Parma University, Italy*
10:15 - 10:30	Real-time cooperative response to light in breathing crystals driven by a photoinduced elastic field	E. Collet *University Rennes, France*
10:30 - 11:00	Coffee Break	

SESSION 17	**FUNCTIONAL MATERIALS - INTRAMOLECULAR PROPERTIES**	
11:00 - 11:30	Using muons to probe order and excitations in molecule-based spin chains and ladders	T. Lancaster *Durham University Physics, Great Britain*
11:30 - 11:45	Mixed stack CT crystals: old materials still holding the scene	A. Girlando *Parma University Chemistry, Italy*
11:45 - 12:00	Stochastic magnetization jumps in chiral molecular magnets	R. Morgunov *Institute of Problems of Chemical Physics, RAS, Russia*
12:00 - 12:10	Closing	Martin Dressel

Oral
Session 1

SUPERCONDUCTIVITY I

Monday, September 7, 2015

09:00 - 10:30

09:00

Molecular layers in iron-based superconductors

S. Blundell
University of Oxford, Department of Physics, Great Britain

Molecular groups can now be intercalated into iron-based superconductors with dramatic consequences on the superconducting properties. These species act as charge reservoirs, sources of electrical polarization, and also make subtle structural modifications to superconducting layers, all of which can make novel adjustments to the band structure that in turn can control superconducting properties. By synthesizing the compound $Li_x(NH_2)y(NH_3)_{1-y}Fe_2Se_2$ (x ~ 0.6; y ~ 0.2), in which lithium ions, lithium amide and ammonia (NH3) act as the spacer layer between FeSe layers, we have turned a 9 K superconductor into a 43 K superconductor [1]. Further chemical modification allow us to produce a range of new superconducting materials which we have studied using a variety of techniques including muon-spin rotation. Recently, we have used hydrothermal reactions to produce layered lithium iron selenide hydroxides with chemical formula $Li_{1-x}Fe_x(OH)Fe_{1-y}Se$ and thereby producing compounds whose transition temperature can be tuned from zero up to about 40 K [2]. Minimizing the concentration of iron vacancies in the iron selenide layer and simultaneously increasing the electron count on iron in the selenide layers enhance the superconducting properties in this family. These new families of materials offer the potential to link the field of molecular superconductors with that of iron-based superconductors and I will discuss future prospects for new superconducting materials using these techniques. (Work performed in collaboration with S. J. Clarke and coworkers at Oxford, RAL and Durham, UK.)

[1] M. Burrard-Lucas et al., Nature Materials 12, 15 (2013).
[2] H. Sun et al. Inorg. Chem. 54, 1958 (2015).

09:30

Magnetism and electron-phonon interaction in the emergence of organic superconductivity

C. Bourbonnais[1], H. Bakrim[2]
[1]*Université de Sherbrooke, Département de physique, Canada*
[2]*n/a, Germany*

In this talk, we analyze the influence of electron-phonon interaction on unconventional superconductivity, as driven by spin fluctuations in correlated quasi-one-dimensional electron systems [1]. We employ the renormalization group method to treat on the same footing the electron-electron and the tight-binding electronphonon interactions for the quasi-one-dimensional electron gas model. We show that for a momentum dependent phonon-mediated interaction that is weak compared to nonretarded repulsive interactions, spin fluctuations and in turn d-wave superconductivity are reinforced. As a function of phonon frequency, the results reveal a positive isotope effect for both anti ferromagnetic and d-wave superconducting orders, as well as for the amplitude of quantum critical spin fluctuations effects in the normal phase. We discuss the impact of these results for organic conductors like the Bechgaard salts, which stand out as a classical example of proximity between anti ferromagnetism and superconductivity.

10:00

Disorder-induced gap in the normal density of states of the organic superconductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br

H. - J. Elmers[1], S. Diehl[1], T. Methfessel[1], J. Müller[2], M. Lang[2], M. Huth[2], M. Jourdan[1]
[1]*Universitaet Mainz, Institut fuer Physik, Germany;*
[2]*Goethe Universität, Physikalisches Institut, Frankfurt, Germany*

Scanning tunneling microscopy and spectroscopy was applied to in-situ cleaved surfaces of the organic superconductor k-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br at a temperature range from 0.5 Tc to 1.2 T_c (T_c denoting the critical temperature). From the differential conductivity we extract the local density of states (DOS) at the surface of the organic superconductor. Data has been acquired for tunneling parallel and perpendicular to the conducting BEDT-TTF planes. The DOS reveals a logarithmic suppression near the Fermi edge persisting above the critical temperature Tc on both surface orientations [1]. The experimentally observed suppression of the DOS is in excellent agreement with a soft Hubbard gap as predicted by the Anderson-Hubbard model for systems with disorder. The electronic disorder also explains the diminished coherence peaks of the quasi-particle DOS below Tc. Dividing the DOS by this temperature independent soft Hubbard gap we obtain the density of states related to the superconducting phase (SC-DOS) [2]. The SC-DOS comprises two separate energy gaps with extrapolated ground state values of D1 = 2.3 meV and D2 = 9.1 meV, both vanishing at Tc. The observed double-gap provides evidence for multiband superconductivity in the title compound. This implies restrictions to possible paring mechanisms. The compatibility of this result with previously published results on thermodynamic properties near Tc is discussed. It should be noted that our interpretation does not cover strong-coupling effects nor does it provide the desirable quantitative description of the energy gaps, requiring further research.

[1] S. Diehl, T. Methfessel, J. Müller, M. Lang, M. Huth, M. Jourdan, and H. J. Elmers, arXiv:1410.5245
[2] S. Diehl, T. Methfessel, J. Müller, M. Lang, M. Huth, M. Jourdan, and H. J. Elmers ,arXiv:1411.3181

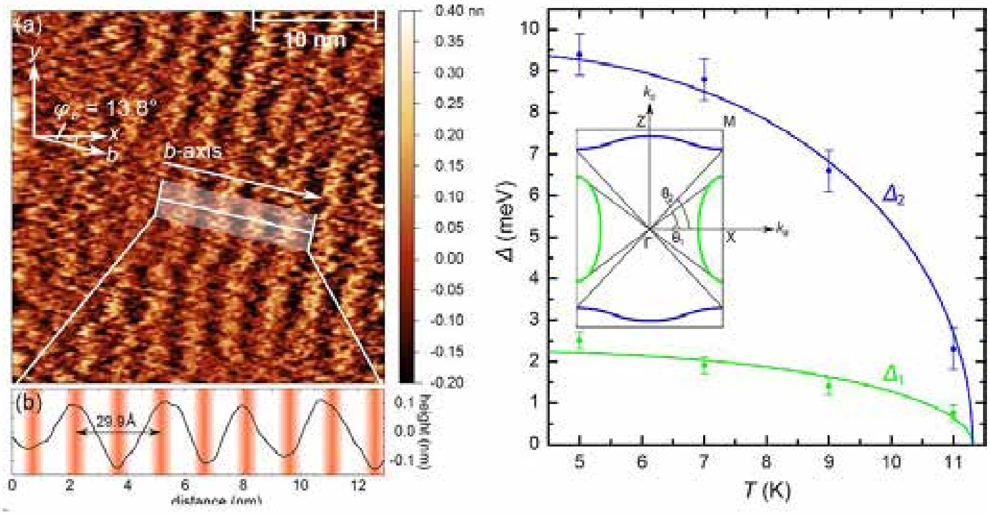


Figure 1: Left: STM image acquired perpendicular to the BEDT-TTF layers, right: SC-DOS gap versus temperature.

10:15

Electron correlation induced superconductivity in the quarter-filled band

R. T. Clay[1] , W. W. de Silva[1], N. Gomes[2], T. Dutta[2], S. Mazumdar[2]
[1]*Mississippi State University, Department of Physics and Astronomy, United States;*
[2]*University of Arizona, Department of Physics, Tucson, United States*

Whether or not electron-electron Coulomb interactions enhance superconducting pair correlations in the weakly doped two-dimensional Mott-Hubbard insulator remains controversial. We present the results of high-precision calculations of superconducting pair-pair correlations on several frustrated lattices over the complete range of carrier density in each case. We find that pair correlations are enhanced relative to the noninteracting limit only for densities ρ at or close to 0.5 (¼ filling). This enhancement is due to the proximity to the spin-gapped paired-electron crystal (PEC) which occurs at ρ=0.5. This proximity also explains the pseudogap observed in many organic superconductors, which is due to preformed pairs in a phase-incoherent paired-electron liquid. We further show results for the full κ-(BEDT-TTF)2X monomer lattice which also show a similar density dependence. The remarkable bandfilling specificity is an essential ingredient to understanding the mechanism of superconductivity in the two-dimensional organic charge-transfer solids as well as the many different families of other unconventional superconductors that share this bandfilling.

This work was supported by US DOE grant DE-FG02-06ER46315. Part of the numerical calculations were supported by the National Energy Research Scientific Computing Center (NERSC) under the US DOE Contract No. DE-AC02-05CH11231.

Oral
Session 2

DESIGN OF NEW MATERIALS I

Monday, September 7, 2015

11:00 - 12:30

11:00

Controlling crystallographic and electronic structure in cationic and anionic radical salts

J. A. Schlueter[1,2]
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The manners by which molecules arrange themselves in the solid state have significant implications for the electronic and magnetic properties of the resultant material. Small energy differences between polymorphic structures can enable crystallization conditions to dictate the phase of the crystallized product. In many cases, competition for directional intermolecular interactions, including hydrogen and halogen bonds, dictates these crystallization pathways. Organic anions represent a huge, under-investigated resource for discovery of new materials. Organic chemistry provides a means to strategically alter chemical structure and tweak intermolecular interactions to probe phase stability and correlated electronic behavior. One particularly promising class of materials involves sulfonate based anions in which both aromatic and aliphatic derivatives are possible. Fluoride decoration of these materials can alter hydrogen bonding patterns. Indeed, it has been shown that this class of materials can stabilize superconductive or magnetic ground states. The second portion of this talk will address new anionic radical salts derived from reductively doped acenes. While this class of materials has potential for superconductivity at temperatures above 25 K, material preparation has been challenging. Through single crystal growth of alkali metal salts it is shown that the choice of alkali metal cation can significantly alter the solid state structure.

This work was supported by the Independent Research and Development program while serving at the National Science Foundation and by Argonne National Laboratory, a U.S. Department of Energy Office of Science laboratory, operated under Contract No. DE-AC02-06CH11357.

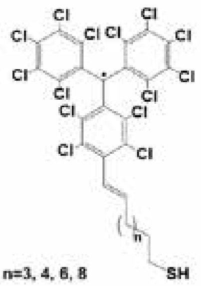
11:30

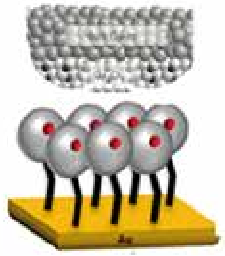
Intramolecular charge transport through organic radicals

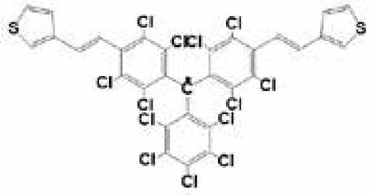
J. Veciana

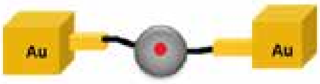
Institut Ciencia Materials Barcelona (CSIC)/CIBER-BBN, Molecular Nanoscience, 08193, Spain

Organic free radicals are neutral molecules exhibiting magnetic properties due to the presence of an unpaired electron. This property together with their low spin-orbit couplings and weak hyperfine interactions make them good candidates for molecular spintronics insofar the radical character is preserved in solid state. In the present contribution, the charge transport through molecules containing persistent polychlorinated triphenylmethyl (PTM) radicals will be presented. These studies have been made with self-assembled monolayers (SAMs) of PTM derivatives grafted on gold surfaces using conducting AFM or contacting the SAMs with top liquid electrodes of eutectic gallium-indium.[1-3] Also the transport through a single PTM radical molecule with a two- (mechanically-controlled break junction) and three-terminal (electromigrated break junction) solid-state devices will be reported.[4] In all cases the conductance through the radical molecules is enhanced two orders of magnitude due to a resonant tunnelling assisted by one of the MOs of these open-shell molecules. Furthermore, the magnetic property of radicals is manifested by a Kondo anomaly in these measurements.









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[3] M. Mas-Torrent, N. Crivillers, V. Mugnaini, I. Ratera, C. Rovira, J. Veciana, J. Mater. Chem., 19, 1691 (2009)

[4] R. Frisenda, R. Gaudenzi, C. Franco, M. Mas-Torrent, C. Rovira, J. Veciana, S.T. Bromley, E. Burzuri, H.S.J. van der Zant, Nano Lett, 15, 3109 (2015)

12:00

Structural and magnetic properties of thiazyl coordination complexes

K. Preuss
University of Guelph, Chemistry, Canada

The so-called "metal-radical approach" to designing molecule-based magnetic materials takes advantage of strong exchange coupling between the moments of a paramagnetic metal ion and the paramagnetic ligand to which it is coordinated. Typically, the nature of the exchange coupling can be understood and predicted based on a simple orbital-overlap model. Employing paramagnetic ligands to mediate strong and predictable magnetic coupling between metal ion moments is an attractive model for material design. We are interested in extending the metal-radical approach by creating paramagnetic ligands that will also engage in intermolecular interactions, through so-called "pancake bonding", highly directional electrostatic contacts, or weaker van der Waals contacts. Our current work on developing paramagnetic ligands designed using cyclic thiazyl "building blocks" will be presented. This work includes examples of molecule-based materials that exhibit ferromagnetic (FM) ordering,[1] antiferromagnetic (AF) ordering,[2] and reentrant phase transitions,[3] arising from the design principles described above.

[1] manuscript in preparation
[2] Fatila, E. M.; Clérac, R.; Rouzières, M.; Soldatov, D. V.; Jennings, M.; Preuss, K. E. J. Am. Chem. Soc. 2013, 135, 13298.
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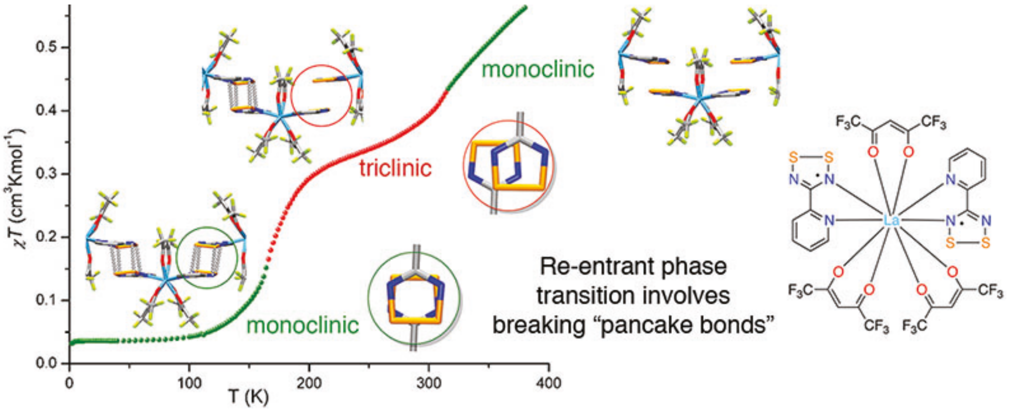

Figure 1: Two sequential phase transitions are observed in the magnetic properties. Although it is not immediately obvious, this is actually a re-entrant phase transition

Oral
Session 3

FUNCTIONAL MATERIALS

Monday, September 7, 2015

14:00 - 16:15

14:00

Intrinsic electrostatic control of TTF-TCNQ molecular layers on noble metal surfaces

P. Maksymovych
Oak Ridge National Laboratory, Center for Nanophase Materials Sciences, United States

Molecular charge-transfer and cation-radical salts continue to be a fascinating source of discoveries and puzzles stemming for strong electron correlations and competing interactions. Our effort pursues low-dimensional epitaxial layers of these compounds, with the focus on understanding their fundamental properties and creating new kinds of metal-organic or organic-organic interfaces [1,2]. One of the primary challenges is to assure that the epilayer is compatible with the redox processes so as to enable, enhance but not eliminate the regime of strong correlations. In this talk, I will discuss the role of electrostatic interactions in 2D molecular structures of TTF-TCNQ supported on metal and graphitic surfaces. We have established that TTF and TCNQ molecules spotnaneously self-assemble into a „zoo“ well-ordered hetero-molecular monolayers and multilayers with a wide-range of donor:acceptor ratio from 2:1 to 1:2 [3] (Fig. 1). Most notably, the terminations of the 2D molecular islands have only very specific, and often counterintuitive structures, including long-range vacancy ordering. From a computational analysis, we conclude that the observed shapes obey minimization of long-range electric fields inside molecular islands, in overall analogy to Tasker's classification of polar terminations of crystalline lattices [4]. Furthermore, electrostatics will have dominant energetic contribution to the kinetics and thermodynamics of epilayers, including their defect structure. In this regard charge-transfer salts are radically different from most homomolecular self-assembled monolayers where electrostatic interactions are almost entirely suppressed. Finally, we will propose that electrostatics is also responsible for formation of decidedly non-bulk like Mott-insulating state in ultrathin multilayers of TTF-TCNQ.

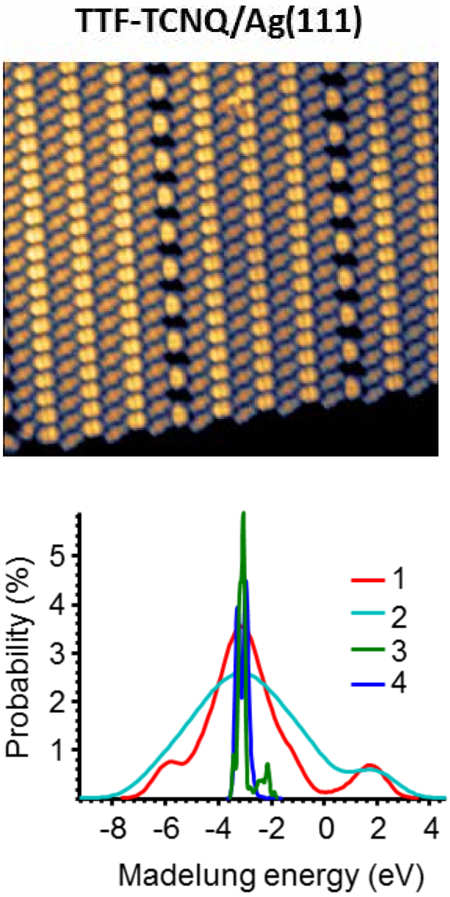

Figure 1: Left: STM image of TTF-TCNQ on Ag(111) showing regular vacancy ordering. Right: probablility density distribution of Madelung energy within four epitaxial structures different only in their edge termination.

This research was conducted at the Center for Nanophase Materials Sciences, which is a DOE Office of Science User Facility.

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14:30

Anilate-based functional molecular materials with conducting and magnetic properties

M. L. Mercuri[1], M. Atzori[1], E. Sessini[1], N. Avarvari[2], A. Serpe[1]
[1]*University of Cagliari, Scienze Chimiche e Geologiche, Monserrato (Cagliari), Italy;*
[2]*Université d'Angers, Laboratoire MOLTECH-Anjou, France*

In the search for new functional molecular materials whose physical properties can be easily tuned by simple changes on the molecular structures of their building units, we have designed and synthesized a family of paramagnetic metal complexes of general formula $[\mathrm{M^{III}(X_2An)_3}]_3$- ($\mathrm{M^{III}}$ = Cr, Fe; X = Cl, Br, I, H; An = 2,5-dihydroxy-1,4-benzoquinone).[1,2] These metal complexes are valuable building blocks for the preparation of functional materials, such as molecule-based ferrimagnets and magnetic molecular conductors, where the nature of the X substituent at the 3,6 positions of the anilate ring can play a key role in determining their physical properties.[3-5] Here we report the synthesis and the full characterization of these metal complexes as well as the synthesis and characterization of a novel family of molecular ferrimagnets formulated as $\mathrm{A[Mn^{II}Cr^{III}(X_2An)_3]}$ (A = $\mathrm{(n\text{-}Bu)_4N}$+, $[\mathrm{(phen)_3(H_3O)}]^+$, X = Cl, Br, I). In this family, a simple change of the substituent atom (X) on the bridging ligand allows for a fine tuning of the magnetic properties: the ordering temperature increases from 5.5 to 6.3, 8.2 and 11.0 K for X = Cl, Br, I, H, respectively (see Figure 1, left hand side).[3] Furthermore, by combining the paramagnetic metal complex $[\mathrm{Fe(Cl_2An)_3}]_3$- with the organic donor BEDT-TTF (bis-ethylenedithio-tetrathiafulvalene) and its chiral derivatives (S,S,S,S)- and (R,R,R,R)-TM-BEDT-TTF via electrocrystallization, two novel families of paramagnetic molecular conductors have been obtained. The crystal structures and the physical properties of these magnetic/conducting hybrid systems will be also reported in this communication.[4,5]

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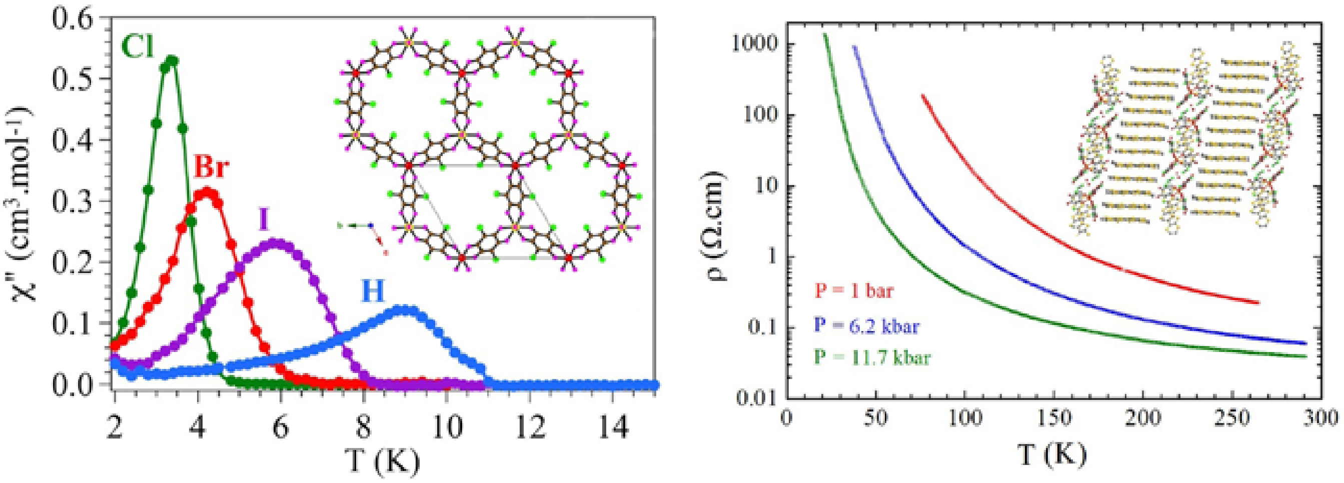

Figure 1: (left) AC susceptibility measurements for the $[\mathrm{Mn^{II}Cr^{III}(X_2An)_3}]$- (X = Cl, Br, I, H) family; (right) Conductivity measurements for the alfa'''-[BEDT-TTF]18 $[\mathrm{Fe(Cl_2An)_3}]_3$•$\mathrm{3CH_2Cl_2}$•$\mathrm{6H_2O}$ system.

15:00

Organic phase-transition transistors with strongly-correlated electrons

H. Yamamoto
Institute for Molecular Science, Research Center of Integrative Molecular Systems, 444-8585, Japan

Direct control of electronic phases and associated physical properties by external stimuli such as electric-field, strain, and light is a key issue in developing new devices for information processors, sensors, and communication infrastructures in the next era. It also helps discovery of unknown electronic phases that are not accessible by static modification such as chemical doping, and thus can contribute to the expansion of fundamental solid state physics. Strongly correlated electron system such as Mott-insulator based on organic materials, where on-site Coulomb repulsion among the carriers can be modulated by both the carrier concentration and the soft lattice spacing, is an excellent playground for demonstrating such a direct switching of electronic phases. Specifically, switching an insulator to a superconductor is one of the most interesting phenomena, because of its large ON/OFF ratio in conductivity, magnetization, and phase coherence. In this presentation, such a direct switching of superconductivity by gate electric field, one-dimensional strain, and/or light-irradiation will be discussed. In case of light, remote control of superconductivity would be also possible.

This work has been done in collaboration with Drs. M. Suda (IMS), Y. Kawasugi, R. Kato (RIKEN), M. Nakano, Y. Iwasa (Univ. Tokyo), T. Minari, and K. Tsukagoshi (NIMS). (BEDT-TTF = bis(ethyelenedithio) tetrathiafulvalene) ref.

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15:30

Conductors and transistors based on BTBT-based charge-transfer complexes

T. Mori[1], T. Kadoya[2], Y. Kiyota[1], T. Higashino[3], K. Iijima[1], M. Dogishi[1], R. Sato[1], T. Kawamoto[1], K. Takimiya[4]
[1]*Tokyo Institute of Technology, Department of Organic and Polymeric Materials, Japan;*
[2]*University of Hyogo, Graduate School of Material Science, Kamigori, Japan;*
[3]*The University of Tokyo, Institute for Solid State Physics, Kashiwa, Japan;*
[4]*RIKEN, Center for Emergent Matter Science, Wako, Japan*

Recently it has been revealed that many small-molecule semiconductors exhibit ambipolar transistor properties, showing both hole and electron transport by using tetratetracontane as the passivation layer. We have shown molecules such as indigo [1], isoindigo, diketopyrrolopyrrole, dicyanomethylene terthiophene [2], and binaphthosemiquinone [3] exhibit excellent ambipolar transistor properties. These molecules are interesting because a single molecule shows both donor and acceptor properties. However, it is still a great challenge to make air-stable electron-transporting transistors [4]. On the other hand, we have found that a charge-transfer salt, $(BTBT)_2PF_6$, shows as high conductivity as 1500 S/cm and metallic temperature dependence [5,6], where BTBT is a famous organic transistor material benzothieno[3,2-b]benzothiophene [7]. This salt exhibits a resistivity jump around 150 K followed by a semiconducting state below 60 K. The AsF_6, SbF_6, and TaF_6 salts are isostructural, having BTBT stacks arranged in a windmill matter. The room-temperature thermoelectric power is about 15μV/K, and $(BTBT)_2AsF_6$ shows a large power factor of $S^2\sigma \sim 55$ μW/m K^2. Tetracyanoquinodimethane (TCNQ) complexes of the BTBT derivatives have mixed-stack columns, but form air-stable n-channel transistors with the mobilities of the 10^{-2} cm^2/Vs order.

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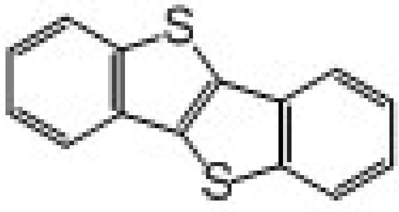

Structure of BTBT

15:45

Preparation of ultrathin films of electrically active Metal-Organic Frameworks

V. Rubio-Giménez, S. Tatay, C. Martí-Gastaldo, E. Coronado
Universitat de València, Instituto de Ciencia Molecular (ICMol), Paterna, Spain

In the last decade, applications for Metal-Organic Frameworks (MOFs) have been found in gas storage, separations, sensing and catalysis. Recently, there is a growing interest in MOFs' optical and electrical properties. However, research regarding their integration in optoelectronic devices is yet to fully blossom.[1] Although the general insulating character of MOFs has limited development, in the last year electrical conductivity has been reported for MOF thick films.[2-4] Investigation of the electrical conductivity of MOF ultrathin films (<100 nm) remains still a challenge due to the synthetic difficulties in producing smooth films with homogenous coverage and low roughness with nanometric accuracy. We report the fabrication of ultrathin films of NASF-1 [5] (below 20 nm) by using a hybrid approach that involves sequential transfer of pre-assembled 2D nanosheets to substrates pre-modified with a Self Assembled Monolayer (SAM) by using the Langmuir-Blodgett (LB) technique. Our Hg-drop electrode junction studies are indicative of electrical conductivity in MOF films with nanometric thickness for the first time and suggest hopping mechanism as the most likely origin.

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16:00

Intramolecular electronic transport studied with organic radicals

C. Rovira
Institut de Ciència de Materials de Barcelona (ICMAB-CSIC)/CIBER-BBN, Nanomol, Cerdanyola, Spain

Mixed-valence (MV) compounds are excellent models for gaining insight into fundamental intramolecular electron transfer (IET) phenomena. Polychlorinated triphenylmethyl radicals (PTM) have in addition to their paramagnetic properties a strong acceptor character that permits to generate their carbanion counterparts at low potential. We have used this property to generate mixed valence radical-anion systems with a variety of bridges and studied the IET phenomena by Electron Paramagnetic Resonance (EPR).[1] EPR has also been used to study the transport through a tetrathiafulvalene bridge by using neutral acceptor redox centres that generate stable radical anions.[2] By other hand, we have also used PTM radicals to go inside the field of *Molecular Spintronics*. Our study relies on the transport properties comparison between two self-assembled monolayers (SAMs) based on polychlorinated triphenylmethyl (PTM) derivatives that exhibit small differences in their molecular structure but strongly differ in their electronic configuration, i.e. closed and open-shell forms. The investigation of the transport properties were performed with PTM radicals with different anchoring bridge on the nature and length. [3] In all cases the open-shell form resulted being significantly more conducting. By density functional calculations, this observation was suggested to occur due to a single-unoccupied orbital (SOMO) mediated transport in the case of the open-shell system. When single molecule measurements are performed at different temperatures, the effect of the free electron is observed. It is shown that the paramagnetism of the PTM radical molecule in the form of a Kondo anomaly is preserved in two- and three-terminal solid-state devices, regardless of the deformation/stretching changes [4].

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[2] a) F. Otón, V. Lloveras, M. Mas-Torrent, et al. Angew. Chem. Int. Ed., 50,10902, 2011. b) J. Calbo, J. Aragó, F. Otón, et al., Chem. Eur. J., 19, 16656, 2013.

[3] a) N. Crivillers, C. Munuera, M. Mas-Torrent, et al.. Adv. Mater., 21, 1177, 2009. b) N. Crivillers, M. Paradinas, M. Mas-Torrent, et al. Chem. Commun., 47, 4664, 2011. c) M. Mas-Torrent, N. Crivillers, V. Mugnaini, et al. J. Mater. Chem., 19, 1691, 2009.

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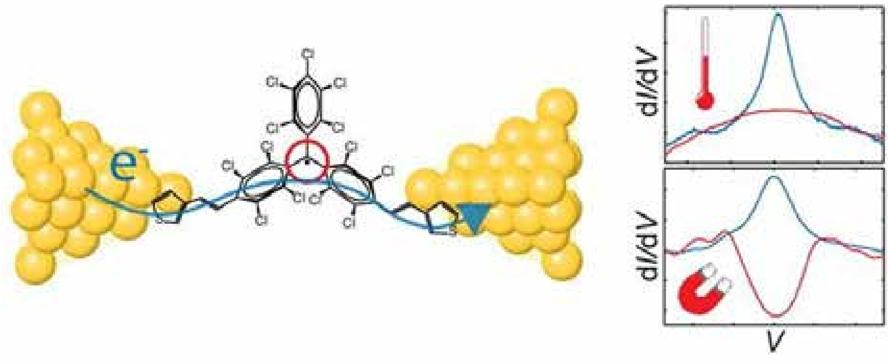

Figure: Kondo resonance on a PTM radical investigated by break junction configutration.

Oral
Session 4

MOTT PHYSICS I

Monday, September 7, 2015

16:45 - 18:00

16:45

Diverse manifestation of electron correlation in molecular materials

K. Kanoda
University of Tokyo, Japan

A many-body quantum system on the verge of instability between competing ground states may exhibit emergent phenomena. Interacting electrons on triangular lattices are likely subjected to multiple instabilities in the charge and spin degrees of freedom. The molecular conductors are superior model systems for studying the issue because of the designability and controllability of the material parameters such as lattice geometry and bandwidth. I present various quantum manifestations that interacting electrons on triangular lattices show under variable correlation, disorder and (unfortunately, fixed) doping in molecular conductors. The topics include i) the quantum criticality of the pressure-induced Mott transition revealed by the resistivity that obeys the material-independent quantum-critical scaling, ii) the emergence of a quantum-disordered spin state from a classical magnet near the Mott-Anderson transition, and iii) a pressure-induced quantum phase transition or crossover from a doped spin-liquid (non-Fermi liquid) to a Fermi liquid and a possible BEC-to-BCS crossover in superconductivity in a doped triangular lattice.

The work presented here was performed in collaboration with T. Furukawa, H. Oike, J. Ibuka, M. Urai, K. Miyagawa (UTokyo), M. Ito, H. Taniguchi (Saitama Univ), R. Kato (RIKEN), M. Saito, S. Iguchi and T. Sasaki (Tohoku Univ).

17:15

Pseudogap phase and the Mott transition in organic and cuprate superconductors

J. Merino
Universidad Autonoma de Madrid, Fisica Teorica de la Materia Condensada, Spain

Organic and cuprate superconductors show a pseudogap phase in a broad region of the phase diagram. The pseudogap phase is evidenced by a suppression of the many-body density of states at the Fermi energy. ARPES experiments in doped cuprates find a suppression in the spectra along the antinodal direction while a quasiparticle-like peak is observed along the nodal direction. Understanding the origin of the pseudogap can be relevant for the understanding of the mechanism of high-Tc superconductivity. In order to understand the origin of the pseudogap phase we have analyzed the single-band Hubbard model in square and anisotropic triangular lattices across the Mott metal-insulator transition.[1,2] Using the dynamical cluster approximation (DCA) on the half-fillied Hubbard model on the square lattice we find that, as the Coulomb repulsion is increased, electrons at the antinodal (pi,0) point open a gap (pseudogap) in the spectral density whereas electrons at the nodal (pi/2,pi/2) point display a quasiparticle-like peak. Our analysis shows how the formation of the pseudogap is associated with the formation of a non-degenerate cluster state and the weaker bath-cluster couplings to the antinodal than to the nodal point. Geometrical frustration and doping effects on the pseudogap are discussed in the context of κ-(BEDT-TTF)$_2$X and cuprate materials.

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17:30

Length changes, critical properties and role of electron-phonon coupling at the Mott transition in κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl

E. Gati[1], R. S. Manna[1], U. Tutsch[1], B. Wolf[1], M. Garst[2], L. Bartosch[3], T. Sasaki[4], H. Schubert[1], J. A. Schlueter[5], M. Lang[1]
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[2]*Universität zu Köln, Institut für Theoretische Physik, Germany;*
[3]*Goethe Universität, Institut für Theoretische Physik, Frankfurt, Germany;*
[4]*Tohoku University, Institute for Materials Research, Sendai, Japan;*
[5]*Argonne National Laboratory, Materials Science Division, United States*

The Mott transition – a key phenomenon of strongly correlated electron systems – and the underlying universality class have been intensively studied from both an experimental and theoretical perspective in the past. The proposal of a hitherto unknown 'unconventional' universality class for the quasi-2D organic charge-transfer salt **κ**-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl ('**κ**-Cl') [1], based on resistivity measurements, has attracted particular interest as it is at odds with considerations suggesting Ising criticality. In this contribution, we report on thermal expansion measurements under ^{4}He gas pressure, a unique novel technique [2], which provides a very sensitive, thermodynamic tool to investigate critical phenomena and related lattice effects [3,4]. It also serves as an ideal probe for studying the effect of electron-phonon-coupling which is supposed to alter the critical properties close to the critical end point by changing the universal behavior to mean-field criticality [5]. For studying such a crossover scenario charge-transfer salts are particularly suited due to their lattice softness. Our study reveals strongly pronounced lattice effects which are highly sensitive to pressure. By analyzing the shift of these effects under pressure within the framework of a scaling theory [4] we show that the thermodynamic response is incompatible with the proposed unconventional universality class. In addition, we will discuss non-linearities in the lattice response as an indication for the effects of strong electron-phonon-coupling in this compound.

[1] F. Kagawa et al., Nature 436, 534 (05).
[2] R.S. Manna et al., Rev. Sci. Instrum. 83, 085111 (12).
[3] M. de Souza et al., PRL 99, 0370031 (07).
[4] L. Bartosch et al., PRL 104, 245701 (10).
[5] M. Zacharias et al., PRL 109, 176401 (12).

17:45

Exploring Mott criticality in κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br by utilizing glasslike structural ordering

B. Hartmann[1], T. Sasaki[2], J. Müller[1]
[1]*Goethe University Frankfurt, Institut of Physics, Germany;*
[2]*Tohoku University, Institute for Materials Research, Sendai, Japan*

We utilize a glass-like structural transition in order to induce a Mott metal-insulator transition in organic charge transfer salt κ-(BEDT-TTF)2Cu[N(CN)$_2$]Br. In this material, the terminal ethylene groups of the BEDT-TTF molecules can adopt two different structural orientations within the crystal structure, namely eclipsed (E) and staggered (S). When cooling through the glass transition at T_g ~ 75 K, a small fraction that depends on the cooling rate remains frozen in the S configuration. We demonstrate that, when thermally coupled to a low-temperature heat bath, a pulsed heating current through the sample causes a very fast relaxation with cooling rates at T_g of order several 1000 K/min [1]. The freezing of the structural orientation causes a decrease of the electronic bandwidth W with increasing cooling rate, and hence a Mott metal-insulator transition as the system crosses the critical ratio $(W/U)_c$ of bandwidth W to on-site Coulomb repulsion U. Due to the glassy character of the transition, the effect is persistent below T_g and can be utilized to fine-tune across the Mott transition by relaxing the sample at Tg (warming cycles, see figure). Comparative measurements in externally applied He-gas pressure may provide a remapping of warming cycles to hydrostatic pressure and allow to distinguish the influence of disorder from the change in bandwidth. By means of this effect, charge carrier dynamics has been investigated in the context of Mott criticality [2].

[1] B. Hartmann, T. Sasaki and J. Müller, Phys. Rev. B 90, 195150 (2014)
[2] B. Hartmann, D. Zielke, J. Polzin, T. Sasaki and J. Müller, Phys Rev. Lett. (in press) (2015)

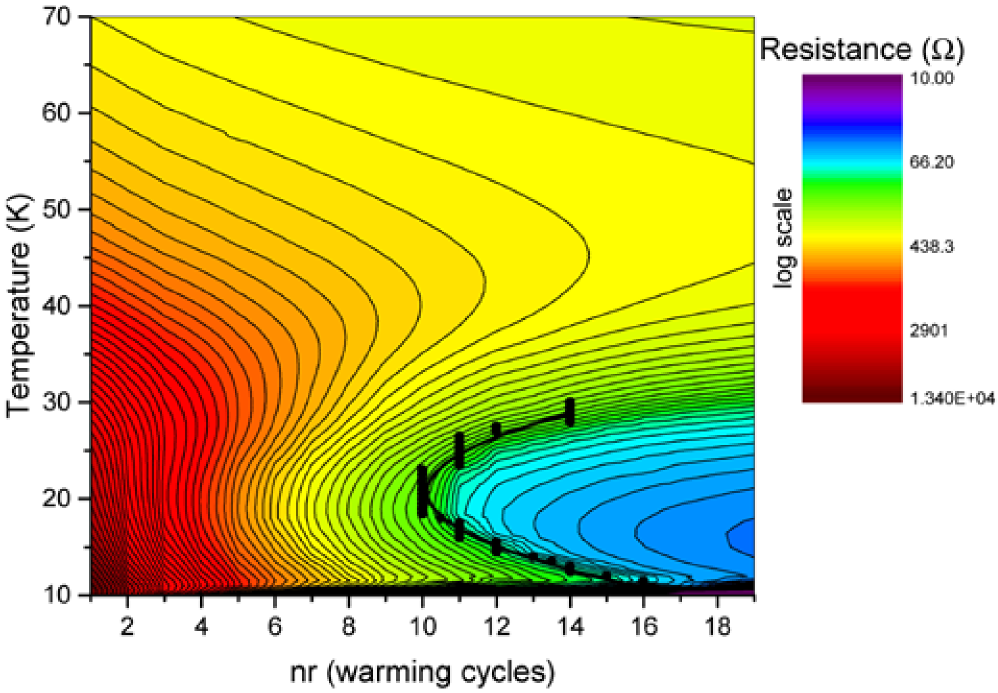

Figure 1: Resistance color plot of κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br after a voltage pulse as a function of temperature and warming cycles. Points are maxima in d(log(R))/d(nr) emphasized by a guide to the eye line representing the first order Mott MIT transition

Oral
Session 5

DIRAC ELECTRON SYSTEMS

Tuesday, September 8, 2015

09:00 - 10:30

09:00

Single-component molecular conductor with Dirac cones

R. Kato[1], T. Tsumuraya[1,2], H. Kino[2], T. Miyazaki[2], H. Cui[1]
[1]*RIKEN, Condensed Molecular Materials Laboratory, Saitama, Japan;*
[2]*National Institute for Materials Science (NIMS), Ibaraki, Japan*

The Dirac cone, where the occupied band contacts with the unoccupied band at a point, is known to induce exotic transport properties. The linear dispersion around the contact point realizes "massless Dirac fermions". Although the Dirac cone is observed in graphene and edge states of topological insulators, the number of the Dirac system in bulk crystals is quite limited. Single-component molecular conductors belong to a multi-band system where both HOMO and LUMO bands contribute to the formation of conduction bands. In most single-component molecular crystals, these two bands are separated from each other at ambient pressure, which leads to insulating properties. Application of high pressure can widen the band width for both bands and their overlap induces the conducting behavior including metallization[1] and superconductivity[2]. In the process of resistivity measurements and the first principles DFT calculations for single-component molecular crystals under high pressure, we noticed a possibility that the (tilted) Dirac cones are formed in a crystal of dithiolene complex [Pd(dddt)$_2$] (dddt = 1,4-dithiin-2,3-dithiolate) which shows temperature-independent resistivity (zero-gap behavior) at 12.6 GPa. The Dirac cone formation in [Pd(dddt)$_2$] can be understood by a simplified two-dimensional tight-binding model where the HOMO band is convex upward and the LUMO band is convex downward. An overlap of these two bands provides a closed intersection at the Fermi level (Fermi line) in the **k**-space, if there is no HOMO-LUMO interaction. An introduction of the HOMO-LUMO interactions removes the degeneracy on the Fermi line and results in a gap formation. However, if there is a line on which the HOMO-LUMO interaction is zero, the two bands contact at the points where the line intersects with the Fermi line, leading to the formation of the Dirac cones.

[1] H. B. Cui, T. Tsumuraya, T. Miyazaki, Y. Okano, and R. Kato, Eur. J. Inorg. Chem., 3837 (2014).
[2] H. B. Cui, H. Kobayashi, S. Ishibashi, M. Sasa, F. Iwase, R. Kato, and A. Kobayashi, J. Am. Chem. Soc., 136, 7619 (2014).

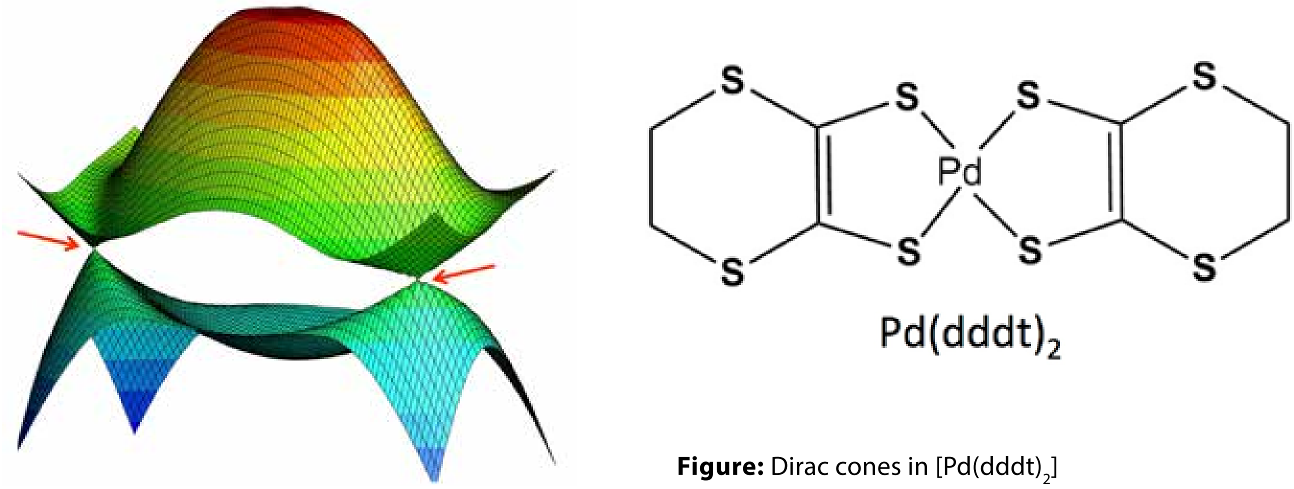

Figure: Dirac cones in [Pd(dddt)$_2$]

09:30

Spin-Order in molecular massless dirac fermion systems

N. Tajima
Toho University, Physics, Funabashi, Japan

First bulk (multilayer) two-dimensional (2D) zero-gap state with massless Dirac particles was realized in an organic conductor α-(BEDT-TTF)$_2$I$_3$ under pressure [1]. We have succeeded in detecting the zero-mode Landau level and its spin-split levels in this system probed by inter-layer magnetoresistance. Recently, Osada pointed out that N=0 quantum Hall ferromagnetism state was realized in this spin-splitting state at low temperature and high magnetic field [2]. In this situation, the interaction between layers plays an important role in the spin textures. In this work, we experimentally demonstrated that the interaction between layers gave rise to peculiar spin-ordered state. The effective g-factor estimated from the transport is abruptly drops from approximately 2 at temperature below 2 K. This result is consistent with the realistic theory by Kubo and Morinari [3]. Moreover, we reveal that this effect also yields the peculiar spin splitting of other Landau levels. The effective g-factors estimated from the spin splitting of N=-1 and -2 Landau levels are much smaller than 2. To our knowledge, this is the first example in Dirac fermion materials. In graphene, for example, large g-value due to Skyrmion excitations was reported [4]. We believe that the suppression of Zeeman effect is one of the characteristic feature of multilayered massless Dirac fermion systems.

[1] K. Kajita, , J. Phys. Soc. Jpn. 83, 072002 (2014).
[2] T. Osada, Phys. Status Solidi B 249, 962 (2012).
[3] K. Kubo and T. Morinari, J. Phys. Soc. Jpn. 83, 033702 (2014).
[4] A. F. Young, et al., NPHYS, 8, 550 (2012).

10:00

Experimental determination of anisotropy of dirac cone and van Hove Singularity in the organic dirac fermion system

T. Osada, A. Mori, M. Sato, K. Uchida
University of Tokyo, Institute for Solid State Physics, Kashiwa, Japan

An organic layered conductor α-(BEDT-TTF)$_2$I$_3$ has the Dirac fermionic electron system under high pressures (P > 1.5 GPa). Although the Dirac feature was predicted by a band calculation, few band calculations have successfully reproduced the zerogap state under ambient pressure in this system. Direct observation of the Dirac band using ARPES or STS is difficult due to high pressure. So, even the qualitative band structure has still been unknown. In this paper, we report the experimental determination of the anisotropy of the Dirac band in α-(BEDT-TTF)$_2$I$_3$ using a magnetotransport method. The interlayer resistance was measured as a function of the azimuthal angle of the magnetic field **B** parallel to conduction layers. Generally, a point on a 2D band with the group velocity parallel to **B** gives the dominant contribution to the interlayer conductivity depending on the thermal distribution of occupancy. So, the anisotropic Dirac cone with the elliptic cross section gives the resistance minimum when **B** is perpendicular to the long axis of the ellipsoid. The angle-dependent patterns at low temperatures show the local minimum at the azimuthal angle of ~ 60 deg. (depending on the pressure) measured from the crystalline **a**-axis to the **b**-axis. It indicates that the long axis of the Dirac cone is tilted by ~ -30 deg. from the **a**-axis. When the temperature is increased, the minimum angle switches from ~ 60 deg. to 0 deg. rapidly around T = 30 ~ 40 K with little pressure dependence. It strongly suggests that a van Hove singularity with huge density of states exists at the energy excited from the Dirac point by 3 ~ 4 meV, and that its crossed equi-energy lines are aligned in the crystalline **b**-axis.

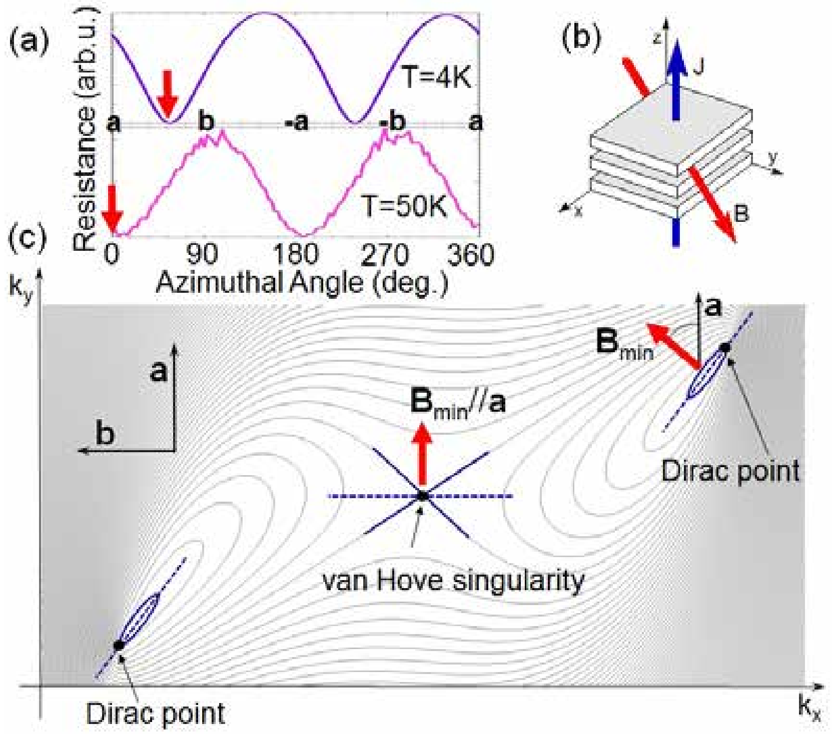

Figure 1:
(a) Azimuthal angle dependence of interlayer resistance.
(b) Experimental configuration.
(c) Schematic configuration of the anisotropic Dirac cones and the van Hove singularity on a 2D band.

10:15

Designing molecular crystals using symmetry and emergence: A study in $Mo_3S_7(dmit)_3$

A. Jacko[1], J. Chander[1], J. Merino[2], B. Powell[1]
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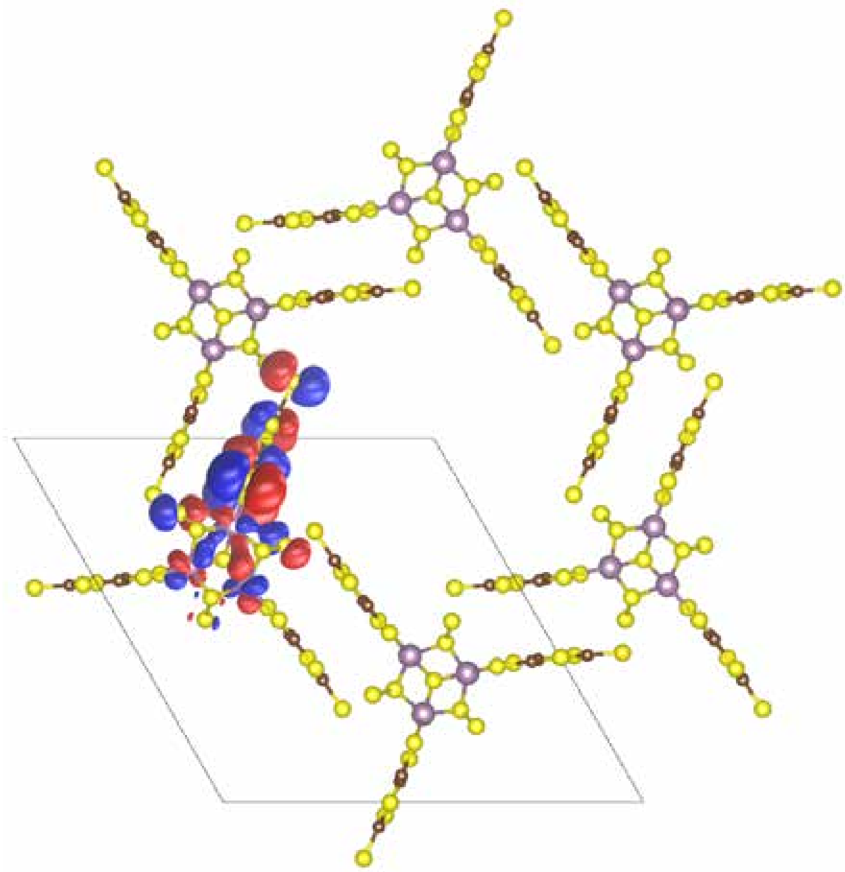
[2]*Universidad Autónoma de Madrid, Departamento de Física Teórica de la Materia Condensada, Spain*

By developing an understanding of the effects of chemistry and symmetry on the emergent physics in certain families of molecular crystals that we can predict the properties of new systems in these families. In this way we can eventually design systems to have specific desired properties. I will present recent work on the spin-liquid candidate $Mo_3S_7(dmit)_3$; building ab *initio* models [1], and describing the emergent properties of this system [2]. $Mo_3S_7(dmit)_3$ is a single component organometallic molecular crystal that was first studied as a potential organic metal. Experimentally, one sees an insulator with well-defined localised magnetic moments, but no long-range magnetic order down to the lowest temperatures measured, hence it is suspected to be a spin-liquid. We report ab *initio* electronic structure calculations that predict the emergence of Dirac fermions and 1D behaviour from a 3D crystal structure due to frustration effects. We discuss the emergence of a '*molecular orbital angular momentum*' as a useful new degree of freedom for understanding relativistic effects in this system. By understanding the emergence of these properties and their connection to symmetry, we can design molecular crystals to exhibit specific desired properties by designing in particular forms of interactions.

[1] A. C. Jacko et al. PRB 91, 125140 (2015) [2] A. C. Jacko et al. In preparation.



$Mo_3S_7(dmit)_3$: Crystal structure of $Mo_3S_7(dmit)_3$, showing the two molecules per unit cell, and one of the 6 Wannier orbitals, localised on a single dmit ligand.

Oral
Session 6

CHARGE ORDER I

Tuesday, September 8, 2015

11:00 - 12:30

11:00

Charge glass in θ-(ET)$_2$X —Emergent phenomena and functionalities

F. Kagawa
RIKEN, Center for Emergent Matter Science (CEMS), Wako, Japan

The ordering kinetics of a first-order phase transition, such as crystallization, consists of two processes—nucleation and growth—and thus requires a finite time to be completed. It follows that crystallization can be kinetically avoided when the temperature range in which the crystallization rate is high is quickly passed through under sufficiently fast cooling. Such kinetic avoidance naturally leads to a nonequilibrium state; a well-established example is supercooled liquids, which generally turn into a glass at low temperatures. In this talk, we show that θ-(ET)$_2$X organic conductors exhibit a glassy state associated with a charge/lattice-configurational degree of freedom only if a first-order charge-ordering transition, or “charge crystallization”, is kinetically avoided [1-3]. The concept of “charge-glass-forming ability” is introduced to understand the phase diagram of θ-(ET)$_2$X [3], and we discuss the relationship between the geometric frustration and the charge-crystallization kinetics. We also demonstrate phase-change-memory (PCM) functionalities emerging from a charge/lattice-configurational degree of freedom in θ-(ET)$_2$X. Repeatable switching between a high-resistivity charge-ordered state and a low-resistivity quenched charge glass is achieved via both optical and electrical heating [4]. The idea of PCM is also applied to an inorganic material with a first-order magnetic transition [5].

[1] F. Kagawa, T. Sato et al., Nat. Phys. 9, 419 (2013).
[2] T. Sato, F. Kagawa et al., Phys. Rev. B 89, 121102(R) (2014).
[3] T. Sato, F. Kagawa et al., J. Phys. Soc. Jpn. 83, 083602 (2014).
[4] H. Oike, F. Kagawa et al., Phys. Rev. B 91, 041101(R) (2015).
[5] H. Oike, F. Kagawa et al., in preparation.

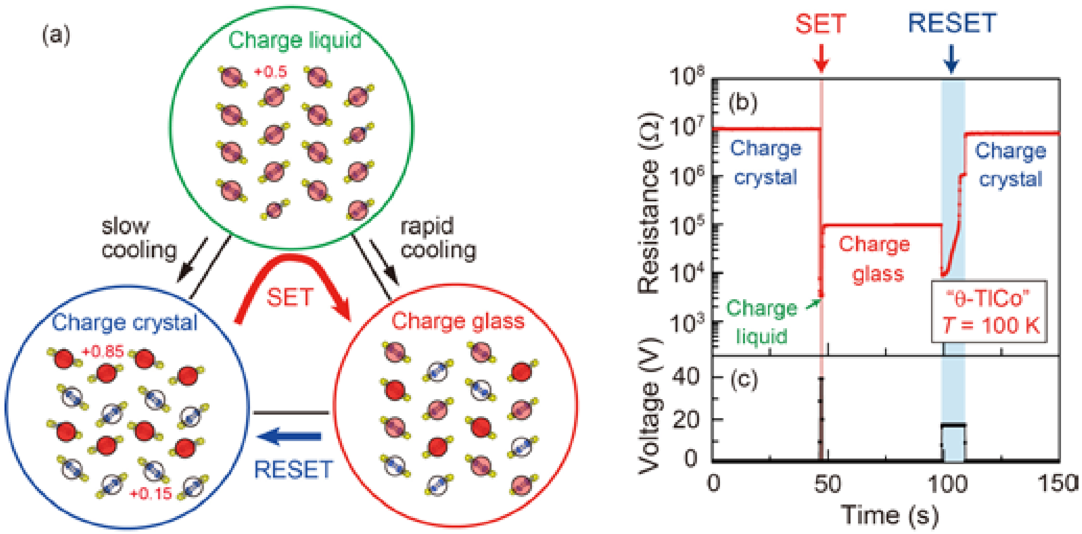


Figure 1: (a) A scheme for the rewritable switching of the correlated-electron phase-change memory implemented using theta-(BEDT-TTF)$_2$X. (b) Single-cycle operation of the SET and RESET processes under an application of rectangular voltage pulses.

11:30

Phase diagram of the triangular extended Hubbard model

L. F. Tocchio[1], C. Gros[2], X. - F. Zhang[3], S. Eggert[3]
[1]*International School for Advanced Studies (SISSA), Trieste, Italy;*
[2]*University of Frankfurt, Institute for Theoretical Physics, Frankfurt a.M., Germany;*
[3]*University of Kaiserslautern, Physics Department and Research Center OPTIMAS, Germany*

We study the extended Hubbard model on the triangular lattice as a function of filling and interaction strength. The complex interplay of kinetic frustration and strong interactions on the triangular lattice leads to exotic phases where long-range charge order, antiferromagnetic order, and metallic conductivity can coexist. Variational Monte Carlo simulations show that three kinds of ordered metallic states are stable as a function of nearest neighbor interaction and filling. The coexistence of conductivity and order is explained by a separation into two functional classes of particles: part of them contributes to the stable order, while the other part forms a partially filled band on the remaining substructure. The relation to charge ordering in organic charge transfer salts is discussed.

Luca F. Tocchio, Claudius Gros, Xue-Feng Zhang, and Sebastian Eggert, Phys. Rev. Lett. 113, 246405 (2014).

12:00

Tilted stripe-type charge order and destabilization of the threefold state in θ-(BEDT-TTF)$_2$X

H. Seo[1], M. Naka[2]
[1]*RIKEN, Condensed Matter Theory Laboratory, Wako, Japan;*
[2]*Tohoku University, Department of Physics, Sendai, Japan*

Charge ordering (CO) with long periods in a geometrically frustrated system, θ-(BEDT-TTF)$_2$X, is theoretically studied [1]. We consider interacting fermion models on an anisotropic triangular lattice: the extended Hubbard model and spinless fermion model, taking account of inter-site Coulomb interactions on not only the nearest-neighbor (NN) bonds but also next-nearest-neighbor (NNN) bonds. By applying the Lanczos exact diagonalization method, we find that the threefold state discussed in the literature found in the charge-frustrated region [2] becomes destabilized when NNN terms are included. Instead, long period charge correlations become dominant, which we interpret as “tilted stripe”-type CO; the resulting state is qualitatively distinct from the threefold CO. The calculated optical excitation spectra show that the charge transfer excitations in the tilted stripe-type CO extend to a lower energy range than those in the previously-discussed CO states, including the threefold CO, stabilized when NNN terms are absent. Our results suggest that the charge correlations experimentally observed in the title compounds near the CO insulating phase [3] can be viewed as a tendency toward a tilted stripe-type CO with large charge fluctuations.

[1] M. Naka and H. Seo, J. Phys. Soc. Jpn. 83, 053706 (2014).
[2] for a review, H. Seo, J. Merino, H. Yoshioka, and M. Ogata, J. Phys. Soc. Jpn. 75, 051009 (2006).
[3] for a review, T. Takahashi, Y. Nogami, and K. Yakushi, J. Phys. Soc. Jpn. 75, 051008 (2006).

12:15

Optical investigation of charge fluctuations in the organic superconductor β''-(BEDT-TTF)$_4$[(H$_3$O)Ga(C$_2$O$_4$)$_3$]C$_6$H$_5$NO$_2$

A. Pustogow[1], Y. Ihara[2], A. Kawamoto[2], M. Dressel[1]
[1]*Universität Stuttgart, 1. Phys. Institut, Germany;*
[2]*Hokkaido University, Department of Physics, Sapporo, Japan*

Charge order is an insulating ground state due to electronic correlations with basically no dc charge transport. Nevertheless, experiments have confirmed theoretical predictions that charge fluctuations (in analogy to spin fluctuations in high-TC superconductors) can be closely linked to superconductivity [1,2]. There are a number of systems which contain charge-ordered insulators and isostructural superconducting compounds next to each other in the phase diagram, e.g. θ-(BEDT-TTF)$_2$I$_3$ [3], β''-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$ [1] and β''-(BEDT-TTF)$_4$[(H$_3$O)M(C$_2$O$_4$)$_3$]Y [4]. In this work we focused on the latter family and investigated the organic superconductor β''-(BEDT-TTF)$_4$[(H$_3$O)Ga(C$_2$O$_4$)$_3$]C$_6$H$_5$NO$_2$ (TC = 7 K). This compound features an extremely high upper critical field H_{c2} = 33 T which makes it a candidate material for the exotic FFLO phase. Transport, Raman [4] (see figure) and NMR [5] studies revealed two charge order transitions at 100 K and 8.5 K, respectively. We performed extended optical experiments covering the FIR and MIR range (50 – 8000 cm^{-1}) for all three polarizations down to low temperatures (T_{min} = 5 K). From out-of-plane measurements (E||c) we determined the degree of charge order. The in-plane response showed rather small anisotropy compared to other organic superconductors, Drude behaviour below 100 cm^{-1} and a broad charge transfer band, indicating a strong influence of charge fluctuations.

[1] S. Kaiser et al., Phys. Rev. Lett. 105 (2010) 206402
[2] J. Merino and R. H. McKenzie, Phys. Rev. Lett. 87 (2001) 237002
[3] H. Mori et al., Phys. Rev. B 57 (1998) 12023
[4] A.F. Bangura et al., Phys. Rev. B 72 (2005) 014543
[5] Y. Ihara et al., Phys. Rev. B 90 (2014) 121106

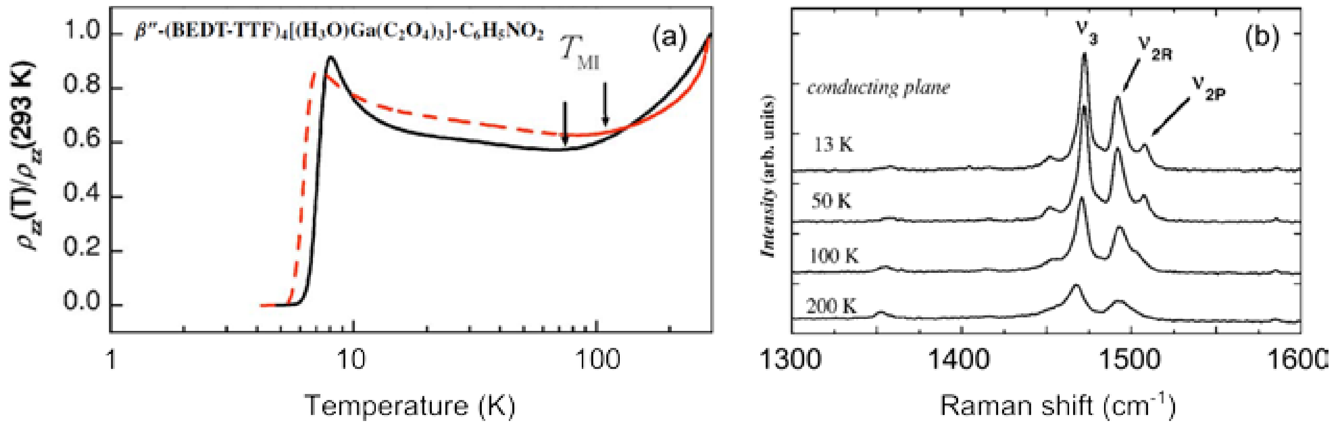


Figure: (a) The upturn of resistivity at 100 K indicates the onset of charge order. (b) Raman investigations disclosed the splitting of a charge-sensitive intramolecular vibration mode and thus confirmed charge disproportionation [4].

Oral
Session 7

QUANTUM SPIN LIQUIDS I

Tuesday, September 8, 2015

14:00 - 15:45

14:00

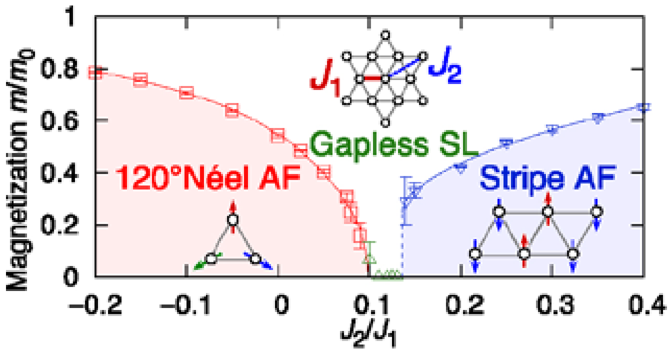
Insight from computational approaches into superconductors and spin liquids in molecular solids

M. Imada
University of Tokyo, Dept. of Applied Physics, Japan

Computational approaches offer powerful tools in revealing nature and mechanisms of various novel quantum phases in strongly correlated electron systems, to which molecular solids belong. Here we focus on two fronts of computational researches on novel quantum phases, one, superconductors and the other, quantum spin liquids, on which organic conductors provide us with a rich variety and their interplay. We first discuss a universal mechanism of superconductivity recently identified in models of cuprate [1] and iron-based [2] superconductors, which is expected to be relevant also in the superconductivity in molecular solids. The mechanism is characterized by electron density fluctuations [1,2] toward inhomogeneity induced by Mottness and emergence of hidden fermions [3], distinct from the boson-mediated superconductivity known in spin-fluctuation as well as BCS-type phonon mediated superconductivity. Candidates of quantum spin liquids are found in several organic Mott insulators. We show liquid-like region sandwiched by two distinct antiferromagnetic orders in the quantum Heisenberg model on square- and triangular lattices with next-nearest neighbour exchange couplings [4,5], which offer simplified models of organic conductors. We also discuss the role of multi-scale ab initio numerical method developed for strongly correlated electron systems [6] in clarifying the realistic mechanisms of the two phenomena in molecular solids.

This series of works has been done in collaboration with T. Misawa, S. Sakai, M. Civelli, S. Morita and R. Kaneko.

[1] T. Misawa, and M. Imada: Phys. Rev. B 90, 115137 (2014).
[2] T. Misawa, and M. Imada: Nat. Commun. 5, 5738 (2014)
[3] S. Sakai, M. Civelli and M. Imada: arXiv:1411.4365
[4] R. Kaneko, S. Morita and M. Imada: J. Phys. Soc. Jpn. 83, 093707 (2014).
[5] S. Morita, R. Kaneko and M. Imada: J. Phys. Soc. Jpn. 84, 024720 (2015)
[6] M. Imada and T. Miyake: J. Phys. Soc. Jpn. 79, 112001 (2010).



Candidate of spin liquid: Spin liquid region found in Heisenberg model on triangular lattice with next nearest neighbour exchange [4].

14:30

New quantum spin-liquid candidates, valence-bond solid, and superconductors κ-(BEDT-TTF)$_2$X (X=Ag$_2$(CN)$_3$, B(CN)$_4$, CF$_3$SO$_3$, Ag(CN)[N(CN)$_2$], etc.

G. Saito[1,2], T. Hiramatsu[1], Y. Yoshida[1], A. Otsuka[3], M. Maesato[4], Y. Shimizu[5], H. Ito[6], H. Kishida[6]
[1]*Meijo University, Department of Agriculture, Nagoya, Japan;*
[2]*Toyota Physical and Chemical Research Institute, Aichi, Japan;*
[3]*Kyoto University, Research Center for Low Temperature and Materials Sciences, Japan;*
[4]*Kyoto University, Graduate School of Science, Japan;*
[5]*Nagoya University, Graduate School of Science, Japan;*
[6]*Nagoya University, Graduate School of Engineering, Japan*

Geometrically spin frustrated materials have been one of the main targets in the materials science since theoretically discussed by Anderson in 1973 [1]. The first such system appeared in 2003, namely a quantum spin liquid (QSL) candidate of two-dimensional Mott insulator κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ with a triangular spin-lattice. Based on the geometrical key-keyhole relation between spin (key=a BEDT-TTF dimer) and anion opening (keyhole) in the triangular spin-lattice system κ-(BEDT-TTF)$_2$X, we have prepared new QSL candidates, valence-bond solid, superconductors, antiferromagnets, etc. (X=Ag$_2$(CN)$_3$, B(CN)$_4$, CF$_3$SO$_3$, Ag(CN)[N(CN)$_2$], etc.). The spins of BEDT-TTF dimers exhibit the competition among localization (Mott insulator, antiferromagnet, valence-bond solid), spin frustration (QSL), itinerancy of electrons (metal), and unconventional spin-pairing for superconductivity. We discuss the spin geometry t'/t, electron correlation *U/W*, and the design of the QSL candidates neighboring the metal and superconducting states.

[1] P.W.Anderson, Mater. Res. Bull. 8 153 (1973).
[2] Y.Shimizu, K.Miyagawa, K.Kanoda, M.Maesato, G.Saito, Phys. Rev. Lett. 91, 107001 (2003).

14:45

Quantum Spin Systems in κ-ET salts

M. Maesato[1], S. Yasuhiro[2], O. Akihiro[3], H. Takaaki[4], Y. Yukihiro[4], G. Saito[4,5]
[1]*Kyoto University, Division of Chemistry, Graduate School of Science, Japan;*
[2]*Nagoya University, Department of Physics, Graduate School of Science, Japan;*
[3]*Kyoto University, Research Center for Low Temperature and Materials Sciences, Japan;*
[4]*Meijo University, Department of Agriculture, Nagoya, Japan;*
[5]*Toyota Physical and Chemical Research Institute, Nagakute, Japan*

The first organic spin liquid k-$(ET)_2Cu_2(CN)_3$ has been discovered in a Mott insulator having a nearly regular triangular lattice (t′/t ~ 1) and moderate *U/t*. Here we study the effects of increasing *U/t*, and increasing t′/t, using the new k-ET salts k-$(ET)_2Ag_2(CN)_3$ and k-$(ET)_2B(CN)_4$, respectively. k-$(ET)_2Ag_2(CN)_3$ is a new spin liquid candidate having a nearly regular triangular lattice (t′/t = 0.97). The ^{1}H-NMR and magnetic susceptibility measurements confirmed the absence of magnetic order down to 0.1 K despite the large antiferromagnetic interaction (J ~ 170 K). It also shows a pressure-induced superconducting transition at around 1 GPa, which is larger than that of k-$(ET)_2Cu_2(CN)_3$. In order to study the low energy excitations, we performed heat capacity and magnetic torque measurements. The T-linear specific heat at low temperatures with negligible magnetic field effect, and paramagnetic behavior up to 45 T are observed, indicating the gapless nature of excitations. k-$(ET)_2B(CN)_4$ is a new Mott insulator having a highly distorted quasi-one dimensional triangular lattice. The magnetic susceptibility indeed follows the spin-1/2 Heisenberg antiferromagnetic model with distorted triangular lattice (J′/J = 2, J ~ 120 K). It undergoes a phase transition to a spin gapped ground state at 5 K, which is well below the exchange energy J. Moreover, the spin-lattice relaxation rate shows nearly temperature independent behavior in the wide temperature range, indicating quantum critical behavior. We will also report the high pressure and high magnetic-field effects on k-$(ET)_2B(CN)_4$ to explore exotic phases neighboring to the spin gapped phase.

15:00

Propagating spin excitations in low-dimensional spin liquids investigated using μSR

F. Pratt[1], P. Baker[1,1], S. Blundell[2], T. Lancaster[3], F. Xiao[3], J. Moeller[2,4], Y. Ishii[5], T. Suzuki[5], I. Watanabe[6], R. Kato[6], G. Saito[7]
[1]*STFC, ISIS Muon Group, Didcot, Great Britain;*
[2]*Oxford University, Physics, Great Britain;*
[3]*University of Durham, Department of Physics, Great Britain;*
[4]*ETHZ, Solid State Physics, Zurich, Switzerland;*
[5]*Shibaura Institute of Technology, Department of Physics, Saitama, Japan;*
[6]*RIKEN, Wakoshi Campus, Japan;*
[7]*Meijo University, Faculty of Agriculture, Nagoya, Great Britain*

Propagating spin excitations in quantum disordered magnets such as 1D spin chains or 2D quantum spin liquids (QSL) can provide important information about their underlying nature. Local probe techniques for measuring spin dynamics such as NMR or μSR provide a means to extract the spectral density of fluctuations associated with the spin motion via field dependence of the relaxation rate of the local spin probe. This technique is relatively well established for the case of Heisenberg antiferromagnetic spin chains, such as the organic system DEOCC-TCNQF$_4$, where 1D diffusive motion of spinons was observed using longitudinal field (LF) μSR [1]. Similar behaviour was also observed recently in the copper chain coordination polymer system Cu(pyz)(NO$_3$)$_2$ [2]. The μSR spin dynamics method can be extended from 1D chain systems to 2D molecular QSL systems such as κ-(ET)$_2$Cu$_2$(CN)$_3$. Diffusion in 2D follows a logarithmic field dependence that is significantly different from the power law that applies in 1D. In the case of κ-(ET)$_2$Cu$_2$(CN)$_3$, the LF μSR measurements follow the field dependence for a 2D system and give a 2D diffusion rate that increases with temperature following a weak power law. The experimental results can be compared with two theoretical predictions that have been made for spinon transport in such a QSL. One model is based on Fermionic spinons with a finite Fermi surface [3] and the other model assumes a quantum critical regime for the spinon dynamics [4]. Both models predict a weak power law for the diffusion rate, however only the quantum critical model is consistent with both the sign and magnitude of the power law observed in the μSR data. The μSR spin dynamics method has also been applied to the system Et$_3$MeSb[Pd(dmit)$_2$]$_2$, another example of a molecular 2D QSL. The surprising new result here is that a 1D diffusion model describes the data much better than a 2D model.

[1] F.L. Pratt et al, Phys. Rev. Lett. 96, 247203 (2006).
[2] F. Xiao et al, Phys. Rev. B (in press)
[3] C.P. Nave and P.A. Lee, Phys. Rev. B 76, 235124 (2007).
[4] Y. Qi, C. Xu and S. Sachdev, Phys. Rev. Lett. 102, 176401 (2009).

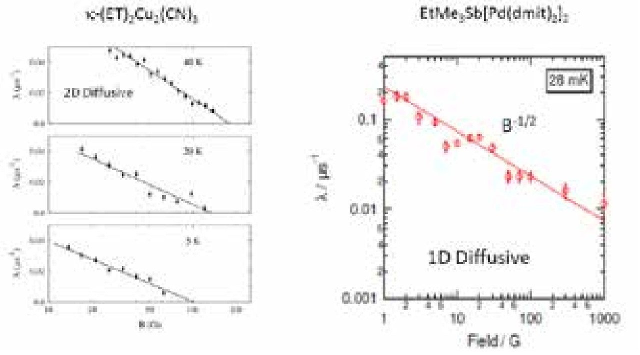


Figure 1: Comparison of the magnetic field dependent muon spin relaxation for two molecular QSL systems

15:15

Ambient pressure superconductivity in nanopatterned crystals of quantum spin liquid κ-$(ET)_2Cu_2(CN)_3$

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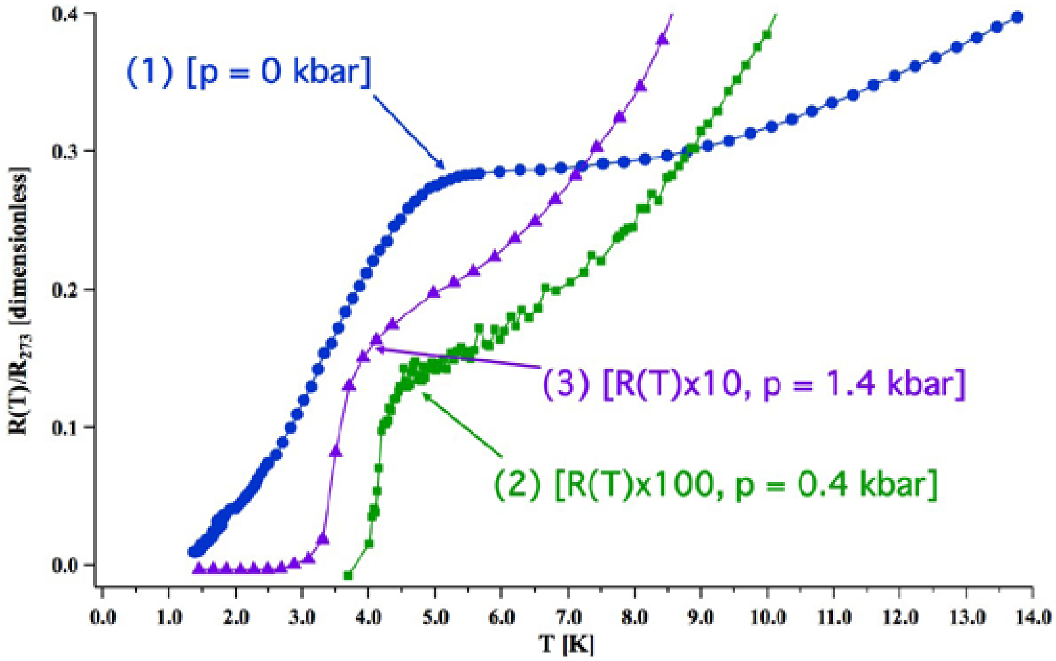
[3]*Institute of Solid State Physics, Russian Academy of Sciences, Chernogolovka, Russia*

Single crystals of quantum spin liquid compound – κ-$ET_2Cu_2(CN)_3$ (κ-CN) were grown by a special technique that includes employing of complex solvent mixtures and high current density conditions. Modulation of growing conditions yielded crystals demonstrated different properties: ambient pressure superconductivity (1) or low pressure superconductivity (2), compared with normal κ-CN phase (3) that has T_c^{on} = 4.2 K at 1.4 kbar. Phase (1) is characterized by a very broad transition and the highest T_c^{on} = 5.5 K. Phase (2) shows very narrow transition and the value of T_c^{on} = 4.5 K at 0.4 kbar that is intermediate between (1) and (3) (see figure). X-Ray and microscopy investigations revealed slightly disoriented mosaic of nanodomains those construct the crystals. No difference in lattice volume and atomic coordinates with the original κ-CN structure was observed. CN group is disordered and positions of C and N atoms are completely intermixed. However, occupancy factors for the positions of CN group tend to vary depending from the size of the crystal under investigation. That hints us that the possible mechanism of superconductivity enhancement is a destruction of ferroelectric domains and thus reducing of the local electric field created by the domains. This is in a contrast with an ambient pressure superconductivity attained by a cell compression that takes place in the case of reported salt with a variable composition – κ'-$ET_2Cu[Cu_xN_{1-x}(CN)_2]CN$ (κ'-CN).



R(T) near SC for (1-3): Nanopatterned crystals of κ-CN demonstrating ambient pressure superconductivity (1) and low pressure (0.4 kbar) superconductivity (2). (3) - Data for normal (insulating at p = 0) crystals of κ-CN at 1.4 kbar. Data for (2) and (3) are scaled by 1

15:30

Dual-functional molecular crystal from candidate quantum spin liquid with Jahn-Teller Distortion

B. Zhang, D. Zhu
Organic Solid Laboratory, Institute of Chemistry, Chinese Academy of Sciences, Beijing, China

Two-dimensional (6,3) and (4,4) Cu lattice, which Cu(II) was octahedral coordinated by format or oxalate anions, were designed as a combination of Jahn-Teller distortion and Resonating valance-bond state. More than thirty ammonium salts of Cu-oxalate were obtained from solution with dimensionanlity from 0D, 1D, 2D to 3D. Ferromagnetic and antiferromagnetic behaviors were observed in 1D, 2D, and 3D system. Ammonium was replaced by organic molecules with p-electron as TTF and its derivatives successfully. The charge-transfer salts q^{21}-(BEDT-TTF)$_3$Cu$_2$(C$_2$O4)$_3$(CH$_3$OH)$_2$, q^{21}-(BEDT-TTF)$_3$Cu$_2$(C$_2$O4)$_3$(C$_2$H$_5$OH)$_2$, and q^{21}-BETS$_3$Cu$_2$(C$_2$O4)$_3$(CH$_3$OH)$_2$ were obtained by electrocrystallization, Donor arrangement and (6,3) lattice anion remained same. 1 and 2 are semiconductor, 3 shows metallic behavior from room-temperature to 180 K. The metallic, semiconductivity charge-transfer salts of (4,4) grid of Cu(II) were obtained when oxalate or format were used as coordination ligands. No long-range magnetic ordering was observed in ammonium and charge-transfer salts with extended Cu-oxalate-framework, and it is confirmed by heat-capacity and single crystal X-ray measurement. They maybe candidate quantum spin liquid.

(BEDT-TTF)$_3$[Cu$_2$(C$_2$O$_4$)$_3$] (CH$_3$OH)$_2$	BETS$_3$[Cu$_2$(C$_2$O$_4$)$_3$] (CH$_3$OH)$_2$	(BEDT-TTF)$_3$[Cu$_2$(C$_2$O$_4$)$_3$] (C$_2$H$_5$OH)$_2$
a = 8.7501(3) Å	a = 8.7716(3) Å	a = 8.8661(5) Å
b = 16.9319(6) Å	b = 16.9903(6) Å	b = 16.7819(8) Å
c = 20.1421(7) Å	c = 20.5716(7) Å	c = 19.4803(9) Å
a = 105.126(1)°	a = 104.794(2)°	a = 74.271(4)°
b = 102.346(2)°	b = 102.209(2)°	b = 88.962(4) °
g = 91.879(1)°	g = 92.243(2)°	g = 88.022(4)°
V = 2801.7(2) Å^3	V = 2883.1(2)Å3	V = 2788.2(2)Å3
290K, P1	290K, P1	290K, P1
Semi, srt = 4 s/cm	Metal to 180K	Semi, srt = 10 s/cm

This work was supported by NSFC 21173230, 973 projects: 2011CB932302, 2013CB933403, and XDB 12030100, P. R. China.

Oral
Session 8

MAGNETIC FIELD EFFECTS

Tuesday, September 8, 2015

16:15 - 17:45

16:15

Hall resistance anomalies in two dimensional organic conductor α-(BEDT-TTF)$_2$KHg(SCN)$_4$

S. Uji[1,2], K. Sugii[3], K. Kobayashi[4], T. Isono[1], T. Terashima[1], J. Yamada[5]
[1]*National Institute for Materials Science, Tsukuba, Japan;*
[2]*University of Tsukuba, Graduate School of Pure and Applied Science, Japan;*
[3]*University of Tokyo, Institute for Solid State Physics, Kashiwa, Japan;*
[4]*Okayama University, Japan;*
[5]*University of Hyogo, Graduate School of Material Science, Ako-gun, Japan*

In layered organic conductucotors, various fascinating transport properties have been found, due to the low dimensionality of the electronic state. For TMTSF-based quasi-one-dimensional conductors, the interlayer resistance at low temperatures in high magetic fields shows unconventional field angular dependece associated with sharp dips at so-called Lebed magic angles (MAs) [1], which is not consistent with Boltzman transport theory (BTT). In addition, the sign change of the giant Nernst effect [2] and Hall resistance [3] is found at the MAs. These features suggests that the current is locked-in the MA layer but the detailed mechansim has not been clarified yet. In order to further investigate such properties, we have measured the resistance of a two dimensional organic conductor α-(BED-TTF)$_2$KHg(SCN)$_4$ in a wide temperature and field region. α-(BED-TTF)$_2$KHg(SCN)$_4$ shows a density-wave (DW) transition at about 8 K (T_{DW}). In the DW phase, the anomalous angular dependent magnetoresistace with sharp dips is observed, which is very similart to that of the TMTSF salt. Figure 1 presents the Hall resistance (R_{xy}) at 1.7 K as a function of θ, which is the field angle from the axis perepndicular to the layer. The BTT predicts that R_{xy} is given by a smooth function, cos(θ) dependece, for layered systems, but the R_{xy} curves suprisingly show sharp features at the MAs. Such featrurs disappear above T_{DW}. The slope of the R_{xy} curve at the MA gives the effective number of the carriers, which is almost field independent. The detailed results will be discussed in terms of the carriers locked in each MA layer and incoherent interlayer transport induced by the field.

[1] E. I. Chashechkina and P. M. Chaikin, Phys. Rev. Lett. 80 (1998) 2181
[2] W. Wu, I. J. Lee, and P. M. Chaikin, Phys. Rev. Lett. 91 (2003) 056601
[3] K. Kobayashi et al., Phys. Rev. Lett. 112 (2014) 11680

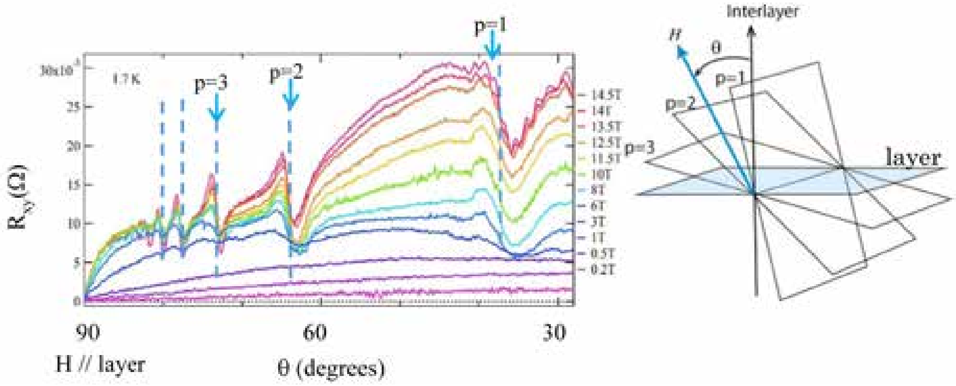


Figure 1: Hall resistance (Rxy) as a function of the field angle and schematic picture of the layers where the carriers are locked in by the field.

16:45

Interlayer charge transport and dimensional crossovers in a layered organic conductor

M. Kartsovnik[1], W. Biberacher[1], P. Grigoriev[2]
[1]*Walther-Meißner-Institut, Bayerische Akademie der Wissenschaften, Garching, Germany;*
[2]*L. D. Landau Institute for Theoretical Physics, Russian Academy of Sciences, Chernogolovka, Russia*

The mechanism of interlayer charge transport has been one of highly debated topics in the field of strongly anisotropic conductors such as organic and metal oxide superconductors. Here we present our understanding of this issue, illustrating it on the behavior of the layered conductor α-(BEDT-TTF)$_2$KHg(SCN)$_4$ pressurized to the normal metallic state. This compound is known for its high anisotropy, $t_{interlayer}/t_{intralayer} \sim 10^{-3}$, relatively simple and well characterized Fermi surface and high crystal quality. We argue that the mechanism of the interlayer transport is unaffected by impurity scattering: the coherent conduction channel remains unchanged irrespective of whether the scattering rate is higher or lower than the characteristic interlayer hopping rate $t_h \sim h/ t_{interlayer}$. In this respect, we find no difference between the fully coherent and so-called „weakly incoherent" transport regimes. An inplane magnetic field has essentially no impact on the charge dynamics but may shift the relative contributions of the coherent and incoherent conduction channels in favor of the latter. By contrast, an out-of-plane magnetic field is found to cause a crossover in the charge dynamics, leading to a field-dependent transport scattering time and modifying the Landau level shape. This is reflected in the shape of the classical magnetoresistance as well as in the harmonic ratio and field dependence of the amplitude of magnetic quantum oscillations.

17:00

Spin–charge coupling in a series of π–d molecular conductors $(DIETSe)_2FeBr_{4x}Cl_{4(1-x)}$

G. Kawaguchi[1], M. Maesato[1], T. Komatsu[1], H. Kitagawa[1,2], T. Imakubo[3], D. Graf[4], A. Kiswandhi[4], J. Brooks[4]
[1]*Kyoto University, Division of Chemistry, Graduate School of Science, Japan;*
[2]*JST-CREST, Tokyo, Japan;*
[3]*Nagaoka University of Technology, Department of Materials Science and Technology, Niigata, Japan;*
[4]*National High Magnetic Field Laboratory, Tallahassee, United States*

The quasi-one-dimensional (Q1D) molecular conductor $(DIETSe)_2FeCl_4$ exhibits a spin-density-wave (SDW) transition at about 12 K, which is followed by an antiferromagnetic (AF) ordering of d-spins of Fe^{3+} at 2.5 K. On the other hand, $(DIETSe)_2FeBr_4$ shows a metal–insulator transition coupled with AF ordering of d-spins at 7 K. Thus, the $FeCl_4$ and $FeBr_4$ salts show much different physical properties. To systematically control the electronic states, we synthesized a series of isostructural mixed-anion salts $(DIETSe)_2FeBr_{4x}Cl_{4(1-x)}$. The crystal structure is robust against random halogen substitution due to iodine bonds between donor and anion. High quality of the mixed salt was confirmed by the observations of quantum oscillations in magnetoresistance (MR) at high magnetic fields. With increasing Br, SDW transition is suppressed and Néel temperature TN increases, suggesting systematic control of dimensionality and π–d interaction. We have investigated the electronic states of mixed salts under high pressure and high magnetic field up to 35 T. The SDW phase is suppressed by applying pressure. The critical pressure to suppress SDW decreases with Br content, suggesting chemical pressure effect. Above the critical pressure, we observed step-like MR anomalies, attributable to field-induced spin-density-wave (FISDW) transitions. The large MR is observed below the AF phase boundary, which shifts to higher field with increasing Br content. Remarkably, the $FeBr_3Cl$ salt with a wide AF phase exhibits unprecedented AF-to-FISDW transition, which is first realized by fine-tuning using chemical substitution. We also discovered mixing-induced magnetic hysteresis unseen in the pristine salts, possibly due to random magnetic exchanges and glassy nature of SDW states.

This work is dedicated to the memory of Prof. Dr. James S. Brooks.

17:15

Electronic states of the organic superconductor $(TMTSF)_2FSO_3$ studied by the Hall effect measurements

W. Kang
Ewha Womans University, Physics, Seoul, Korea Republic (South)

Most of the quasi-one-dimensional organic conductors based on the TMTTF or TMTSF molecules can be categorized in a generic pressure-temperature phase diagram, which allows us to explore the whole one-dimensional physics by changing the anions. However, a few exceptions such as $(TMTSF)_2NO_3$ and $(TMTSF)_2FSO_3$ show substantially different behavior which cannot be fit into such phase diagram. $(TMTSF)_2FSO_3$ is special in a sense that the FSO_3 anions have a finite electric dipole moment in addition to non-centrosymmetric tetrahedral shape. $(TMTSF)_2FSO_3$ undergoes numerous phase changes on cooling under various pressures. Some of them are related with the formation of superstructure of tetrahedral anions, but the others remain largely unexplained. Superconducting transition temperature was unusually high, and most strikingly, angular dependent magnetoresistance showed Yamaji oscillations, the characteristic behavior of corrugated quasi-two-dimensional Fermi surface. This signifies that the electron structures of FSO_3 salt under certain circumstances is substantially different from those of others. While the previous phase diagram of $(TMTSF)_2FSO_3$ was established from the temperature dependence of electrical resistance and thermoelectric power, we studied temperature and pressure dependence (also electric and magnetic field dependence and cooling rate dependence in some cases) of the Hall effect which is very sensitive to the change of carrier concentration and nature of those carriers. We will report new findings which have not been elucidated by longitudinal electron transport measurements alone.

[1] Y. J. Jo, E. S. Choi, Haeyong Kang, W. Kang, I. S. Seo and O. H. Chung, Phys. Rev. B 67, 014516 (2003).
[2] W. Kang, O. H. Chung, Y. J. Jo, Haeyong Kang, I. S. Seo, Phys. Rev. B 68, 073101 (2003).
[3] T. Takahashi, Physica B: Condensed Matter 407, 1757 (2012) This work was supported by the National Research Foundation of Korea(NRF) grant funded by the Korea government (MSIP) (No. 2015001948).

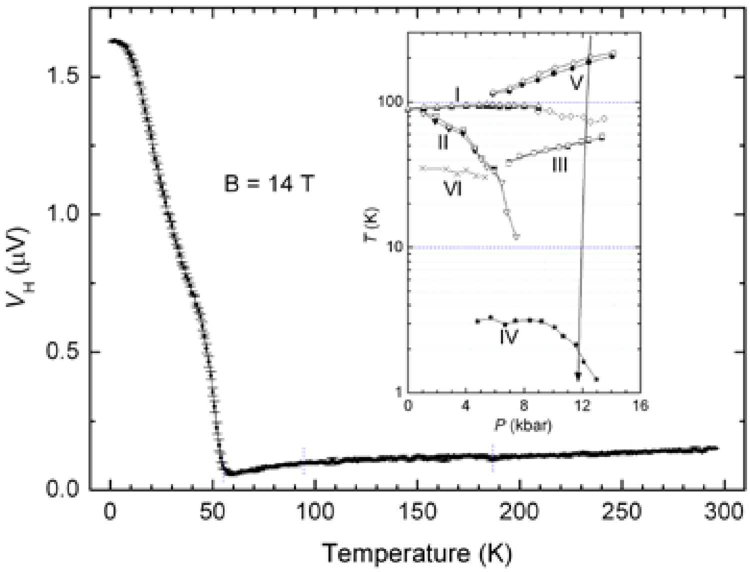


Figure 1: Temperature dependence of the Hall voltage of $(TMTSF)_2FSO_3$ under pressure. Inset shows the T-P phase diagram on which the cooling path of the current measurement is traced.

17:30

Search for the superconducting gap structure in κ-(BEDT-TTF)$_2$X by use of thermal-conductivity measurements

E. L. Green[1], S. Kühlmorgen[1], R. Schönemann[1], J. Müller[2], J. Wosnitza[1]
[1]*Hochfeld-Magnetlabor Dresden (HLD), Helmholtz-Zentrum Dresden-Rossendorf, 01328, Germany;*
[2]*Physikalisches Institut, Goethe-Universität, Frankfurt am Main, Germany*

κ-ET$_2$X, where ET is bis(ethylenedithio)-tetrathiafulvalene (BEDT-TTF), is a family of organic charge transfer salts consisting of conducting ET layers interspersed between insulating X- anions. For almost two decades it has been debated whether these quasi-two-dimensional (Q2D) superconductors are conventional BCS-type or unconventional d-wave similar to the cuprates. A variety of measurements techniques, including NMR, specific heat, and STM, have been employed, but the results do not concur in regards to the pairing symmetry. To investigate this further, thermal conductivity measurements were performed on single crystal X = Cu[N(CN)$_2$]Br and Cu(NCS)$_2$, both of which undergo superconducting transitions at $T_c \approx 11.5$ K and 9.5 K, respectively. In both samples, a relatively large peak in the temperature dependence appears just below T_c which is due to the opening of a superconducting gap and is indicative of high-quality samples. Furthermore, at zero magnetic field, the phonon contribution dominates in both samples at low temperatures (down to 150 mK), as evidenced by a T^3 behavior, with no significant remnant electronic contribution in X = Cu[N(CN)$_2$]Br and only a small contribution in X = Cu(NCS)$_2$. Interestingly, when a magnetic field of μoH = 14 T is applied parallel to the conducting layers, the thermal conductivity is smaller in X = Cu[N(CN)$_2$]Br compared to the zero field data, but is significantly larger in X = Cu(NCS)$_2$. Our results demonstrate the importance of electron-phonon scattering in these Q2D superconductors.

Oral
Session 9

CHARGE ORDER II

Wednesday, September 9, 2015

09:00 - 10:30

09:00

Interplay between structural and electronic effects at the charge ordering transitions of BEDT-TTF and TMTTF quarter-filled organic salts

J. - P. Pouget
Université Paris-sud, Laboratoire de Physique des Solides, ORSAY, France

The physics of many families of two-dimensional (2D) and one dimensional (1D) quarter-filled organic conductors is dominated by the charge localization phenomenon. Although charge localization effects are due to the presence of sizeable electron-electron repulsions, the detailed mechanism of the charge ordering (CO) phase transitions is far to be fully understood. One main reason is that the role of structural degrees of freedom in the CO process is generally underestimated. In this lecture we show that various kinds of structural degrees of freedom play a key role in the CO transition of θ-$(BEDT\text{-}TTF)_2MM'(SCN)_4$ 2D-salts where M= Rb, Cs and M'=Zn, Co, and $(TMTTF)_2X$ 1D-salts, where X is a monovalent anion. In the θ-$(BEDT\text{-}TTF)_2MM'(SCN)_4$ series, DFT electronic structure calculation conjugated to a detailed structural analysis of the interactions between donor and anion layers reveal that that the CO transition in θ-(Rb, Zn and Co) salts is in fact driven by a disorder-order transition of the ethylene-dithio groups of the BEDT-TTF which changes the electronic interactions of the donor layer from 2D to 1D [1]. This reduced dimensionality allows to promote the $4k_F$ charge localization phenomenon typical of correlated 1D quarter-filled electronic systems. In $(TMTTF)_2X$ series, both near-edge X-ray absorption fine structure NEXAFS [2] and Hard X-ray photoemission (HAXPES) [3] measurements allow to probe the role of the anions in the CO symmetry breaking transition of the X=AsF_6 and SbF_6 salts. In addition NEXAFS spectroscopy reveals an enhanced mixing of the AsF_6 anion wave functions with those of the TMTTF when the temperature decreases [2], pointing out for the first time the presence of sizeable anion-donor electronic interactions. Finally we report recent ESR studies of the electrical and magnetic properties of the whole $(TMTTF)_2X$ series which show in particular that CO is extremely sensitive to X-ray irradiation damages [4].

[1] P. Alemany, J.-P. Pouget and E. Canadell, submitted.
[2] K. Medjanik, A. Chernenkaya, S. A. Nepijko, G. Öhrwall, P. Foury-Leylekian, P. Alemany, E. Canadell, G. Schönhense and J.-P. Pouget, submitted.
[3] K. Medjanik, M. de Souza, D. Kutnyakhov, A. Gloskovskii, J. Műller, M. Lang, J.-P. Pouget, P. Foury-Leylekian, A. Moradpour, H.-J. Elmers, and G. Schönhense, Eur. Phys. J. B 87, 256 (2014).
[4] C. Coulon, P. Foury-Leylekian, J.-M. Fabre and J.-P. Pouget, Eur. Phys. J. B 88, 85 (2015).

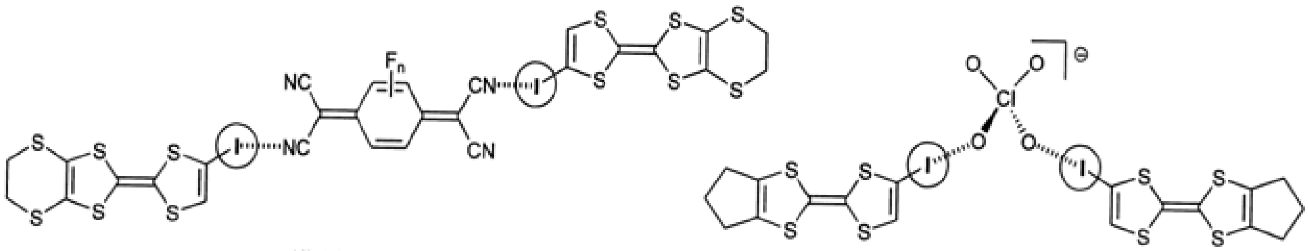
09:30

Charge redistribution in halogen bonded charge-transfer salts of unsymmetrical TTF derivatives studied by IR and Raman spectroscopy

R. Świetlik[1], A. Frąckowiak[1], I. Olejniczak[1], J. Lieffrig[2], O. Jeannin[2], M. Fourmigué[2]
[1]*Polish Academy of Sciences, Institute of Molecular Physics, Poznań, Poland;*
[2]*Universite de Rennes 1, UMR CNRS 6226, Institut des Sciences Chimiques de Rennes, France*

Tetrathiafulvalene (TTF) derivatives substituted with halogen atoms are useful donors for synthesis of charge-transfer (CT) salts since they provide a possibility for a strong, directional interaction with counter ions through halogen bonding. Recently, two 2:1 salts formed by iodinated TTF derivatives with extraordinary properties have been synthesized: **(1)** (EDT-TTFI$_2$)$_2$TCNQF and **(2)** (tTTF-I)$_2$ClO$_4$. In **1** the donors EDT-TTFI$_2$ and acceptors TCNQF are arranged in mixed layers and neighboring layers are liked by C-I···N≡C halogen bond; at room temperature (RT) the salt is essentially neutral and on cooling down it undergoes a neutral-to-ionic transition [1]. On the other hand, in **2** the tTTF-I donors form two types of conducting layers with different molecular arrangement (dual-layer structure with inter-layer I···O halogen bond). In salt **2**, at RT a charge disproportionation is observed which disappears on temperature decreasing; this unique effect is related to an inter-layer CT effect [2]. (FIGURE) Infrared and Raman spectroscopies provided unambiguous proves of the neutral-to-ionic transition in 1 and the interlayer charge disproportionation in **2**. Strong interlayer interactions due to halogen bonding can be at the origin of these effects. Temperature evolution of both electronic and vibrational features as well as the charge redistribution will be discussed.

[1] J. Lieffrig et al. Chem. Eur. J. 19, 14804 (2013)
[2] K.-S. Shin et al. Dalton Trans. 43, 5280 (2014) This work was supported by the Polish National Science Centre (Decision No. DEC-2012/04/M/ST3/00774) and the ANR (France) under contract nb 08-BLAN-0091-02.



(1) (EDT-TTFI$_2$)$_2$TCNQF

(2) (tTTF-I)$_2$ClO$_4$

Halogen bonds: The figure should be put in a place labelled in the text in italics.

10:00

Donor-anion interactions at the charge localization and charge ordering transitions of $(TMTTF)_2AsF_6$ probed by NEXAFS

K. Medjanik[1], A. Chernenkaya[2], S. Nepijko[2], G. Öhrwall[1], P. Foury-Leylekian[3], P. Alemany[4], E. Canadell[5], G. Schönhense[2], J. - P. Pouget[3]
[1] *Lund University, MAX IV Laboratory, Sweden;*
[2] *Johannes Gutenberg-Universität, Institut für Physik, Mainz, Germany;*
[3] *Université Paris-Sud, CNRS UMR 8502, Orsay, France;*
[4] *Universitat de Barcelona, IQTCUB, Spain;*
[5] *Campus de la UAB, ICMAB-CSIC, Bellaterra, Spain*

High-resolution near-edge X-ray absorption fine structure (NEXAFS) measurements at the As M-edge, F K-edge and S L-edge of the Fabre salt $(TMTTF)_2AsF_6$ were performed from room temperature (RT) to 90 K, allowing to reach the charge localization regime below $T\rho \approx 230$ K and to cross the charge ordering (CO) transition at $T_{CO} \approx 102$ K. Upon cooling from RT significant energy shifts up to +0.8 eV and -0.4 eV were observed in transitions exhibited by the F 1s (see Fig.1) and S 2p spectra respectively, while the As 3p doublet does not show a significant shift. Opposite energy shifts found in the F 1s and S 2p spectra reflect substantial thermal changes in the electronic environment of F atoms of the anion and S atoms of TMTTF. The changes found around the charge localization cross-over suggest an increase of the participation of the S d orbitals in the empty states of TMTTF as well as an increase of the strength of donor---anion interactions. A new F 1s pre-edge signal detected upon entry into the CO phase is a clear fingerprint of the symmetry breaking occurring at T_{CO}. We propose that this new transition is caused by a substantial mixing between the HOMO of the AsF_6-anion and the unoccupied part of the TMTTF HOMO conduction band [1].

[1] K. Medjanik et al., submitted to PRB.

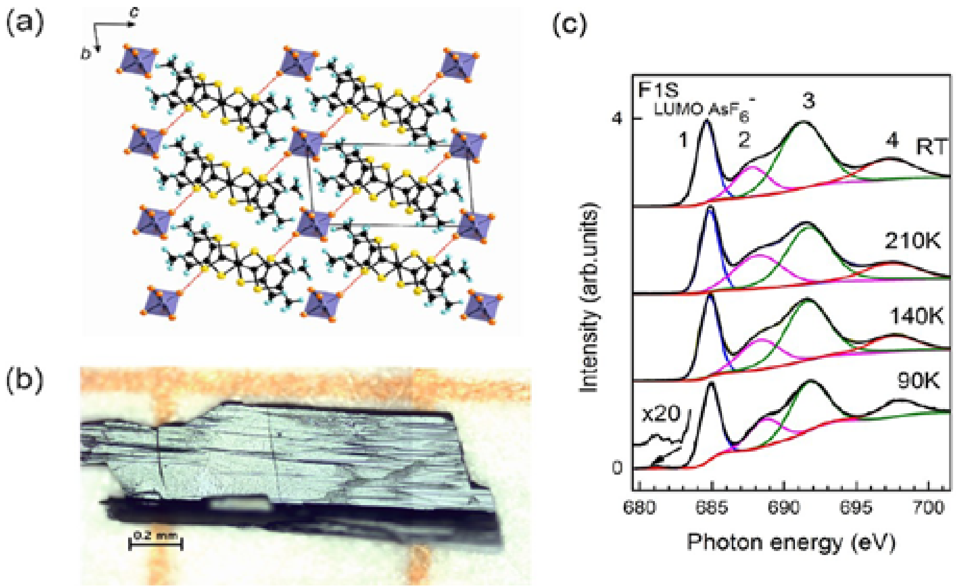


Figure 1: (a) Centro-symmetric P-1 structure of $TMTTF_2AsF_6$ projected along the a stacking direction. (b) Optical image of one (TMTTF)2AsF6 single crystal. (c) F 1s NEXAFS spectra of $(TMTTF)_2AsF_6$ measured at different temperatures.

10:15

Charge degrees of freedom under high pressure in an organic dimer-Mott insulator β’-(BEDT-TTF)$_2$ICl$_2$

K. Hashimoto[1], R. Kobayashi[1], H. Okamura[2], H. Taniguchi[3], Y. Ikemoto[4], T. Moriwaki[4], S. Iguchi[1], M. Naka[5,6], S. Ishihara[5], T. Sasaki[1]
[1]*Tohoku University, Institute for Materials Research, Sendai, Japan;*
[2]*Kobe University, Department of Physics, Japan;*
[3]*Saitama University, Japan;*
[4]*SPring-8, Japan Synchrotron Radiation Research Institute, Sayo, Japan;*
[5]*Tohoku University, Department of Physics, Sendai, Japan;*
[6]*RIKEN, Center for Emergent Matter Science (CEMS), Wako, Japan*

An antiferromagnetic dimer-Mott insulator β’-(BEDT-TTF)$_2$ICl$_2$ exhibits high-temperature superconductivity with T_c = 14.2 K under 8 GPa when the antiferromagnetic phase is suppressed by applying pressure, as seen in other bandwidth-controlled Mott transition systems such as κ-(BEDT-TTF)$_2$X. It has therefore been believed that unconventional superconductivity with dxy-wave symmetry is realized in β’-(BEDT-TTF)$_2$ICl$_2$. However, recent ^{13}C NMR measurements and first-principles band structure calculations have pointed out that the degree of dimerization is significantly reduced with increasing pressure. Recent theoretical calculations based on the extended Hubbard model considering the molecular dimer degree of freedom have revealed that in weakly dimerized systems both charge and spin fluctuations play an essential role in the unconventional superconductivity. Therefore, experimental studies on the electronic structure under high pressure are highly required. Here we report on the pressure evolution of the electronic structure of β’-(BEDT-TTF)$_2$ICl$_2$ via optical conductivity measurements under high pressure. The optical spectra measured at ambient pressure shows that this salt is a typical quasi-1D half-filled dimer-Mott insulator. However, with increasing pressure, the electronic structure of β’-(BEDT-TTF)$_2$ICl$_2$ evolves from the half-filled to 3/4-filled band system, in which the high temperature superconductivity emerges. Our present study highlights the 3/4-filled nature of β’-(BEDT-TTF)$_2$ICl$_2$ under high pressure, indicating that the system approaches the charge-ordered phase with increasing pressure, where charge degrees of freedom inside of dimers become more important. The obtained results suggest that charge fluctuation in addition to antiferromagnetic spin fluctuation plays an important role in the high-T_c superconductivity of β’-(BEDT-TTF)$_2$ICl$_2$.

Oral
Session 10

SUPERCONDUCTIVITY II

Wednesday, September 9, 2015

11:00 - 12:15

11:00

Superconductivity in β''-$(ET)_2SF_5CH_2CF_2SO_3$ at low and high magnetic fields

S. Brown
UCLA, Department of Physics & Astronomy, Los Angeles, United States

The normal and superconducting properties of the highly anisotropic all-organic superconductor (AOSC) superconductor b''-$(ET)_2SF_5CH_2CF_2SO_3$ are probed using ^{13}C NMR. The normal state spin lattice relaxation is contrasted with other organic superconductors. Striking is the absence of evidence for spin fluctuations (SF) in the AOSC, relative to that which is observed for others such as the TMTSF salts In the superconducting state, and which are often associated with the SC pairing mechanism. In addition, there is no evidence for a Hebel-Slichter coherence peak in $1/T_1T$ nor a full gap over the Fermi surface. These results are discussed in the context of the Merino-McKenzie proposal for an alternative superconducting pairing mechanism. At high fields applied parallel to the conducting layers, there is evidence for onset at 9.3(1) T to an inhomogeneous FFLO state in the distribution of spin polarization density inferred from the NMR lineshapes. The transition is evident in the $1/T_1$ as well, and further, there is an indication for a 2nd SC-SC transition at ~11 T.

11:30

Charge density wave-metal (superconductivity) coexistence in the TTF(X(dmit)$_2$)$_2$ family (X=Ni,Pd)

C. Pasquier[1], W. Kaddour[1], P. Auban-Senzier[1], H. Raffy[1], J. - P. Pouget[1], P. Alemany[2], E. Canadell[3], L. Valade[4]
[1]*Laboratoire de Physique des Solides, Bat 510, Centre Universitaire, ORSAY, France;*
[2]*Departament de Quimica Fisica and Institut de Quimica Teorica i Computacional (IQTCUB), Barcelona, Spain;*
[3]*Institut de Ciencia de Materials de Barcelona (CSIC), Campus UAB, Bellaterra, Spain;*
[4]*Laboratoire de Chimie de Coordination, Toulouse, France*

A new pressure-temperature phase diagram of TTF(Ni(dmit)$_2$)$_2$ has been established thanks to spin susceptibility, resistivity and thermopower measurements under pressure up to 30kbar. We were able to find several phase transitions and identify three different charge density wave (CDW) states which all coexist with a metallic state present down to low temperature. This metallicity arises from the development, upon cooling, of a 2D band associated to the Nidmit chains. At low pressure, two successive CDW transitions have been clearly identified and are associated to the successive nesting of two strongly 1D bands: the LUMO and HOMOI of the Nidmit chains. These two transitions merge into a single one at 12kbar which probably corresponds to the partial nesting of a bunch of LUMO's into the HOMOI's. A maximum of this unique CDW transition temperature is observed at 19kbar. The CDW instability associated to the LUMO band is announced by an important regime of Peierls-like fluctuations in the metallic state which give rise to the progressive development of a pseudogap in the spin susceptibility which has been quantitatively analyzed using the Lee-Rice-Anderson theory. The influence of these multiple phase transitions on the superconducting state of TTF(Ni(dmit)$_2$)$_2$ will be also discussed. Finally, a comparison with the phase diagram of the sister system TTF(Pd(dmit)$_2$)$_2$ will be presented.

12:00

Superconductivity beyond the dimer model in single- and multilayered BEDT-TTF based charge transfer salts

H. O. Jeschke, D. Guterding, M. Altmeyer, R. Valentí
Goethe-Universität Frankfurt, Institut für Theoretische Physik, Germany

We theoretically investigate κ-phase BEDT-TTF based charge transfer salts. Based on the experimental crystal structures, we use ab-initio density functional theory to construct realistic models with high accuracy for a broad variety of materials. We analyze the pairing symmetry and strength within random phase approximation spin fluctuation theory. In particular we show that the minimal model for this class of materials needs to describe all BEDT-TTF molecules independently and give an example where the customary dimer model breaks down. Among the materials we investigate, the polymorphs of $(BEDT\text{-}TTF)_2Ag(CF_3)_4TCE)$ (1,1,2-trichloroethane) are of special interest; while a polymorph with a κ packing motif only has low superconducting transition temperature of T_c=2.6K, two high T_c phases are layered structures consisting of α' and κ packed layers. We analzye the influence of the insulating α' layer on the conducting κ layer. Our spin fluctuation theory analysis correctly predicts the trends in superconducting transition temperatures.

Oral
Session 11

FERROELECTRICITY I

Wednesday, September 9, 2015

13:45 - 14:45

13:45

Charge fluctuation and magnetism in dimer Mott insulators

C. Hotta[1], T. Yoshida[2], K. Harada[3]
[1]*University of Tokyo, Basic Sciences, Japan;*
[2]*Kyoto Sangyo University, Physics, Japan;*
[3]*Kyoto University, Graduate school of informatics, Japan*

We deal with a so-called called „dimer Mott insulator" in a classs of two dimensional quarter-filled organic crystal having a dimerized molecular arrangements like kappa- or beta-type. There, every dimer keep one electron localized due to strong electronic correlation. Recent controversy on this dimer Mott insulator is about the origin of the dielectric responses at low enough temperature;the story started from the observation of the relaxor ferroelectric like frequency dependent dielectric anomaly near 6K in kappa-ET_2 $Cu_2(CN)_3$[1].Similar reports followed in other dimer Mott insulating materials like beta'-ET_2I_3[2], dmit compounds [3], and beta-(meso-DMBEDT-TTF)$_2$$PF_6$[4]. Even a seemingly ferroelectric-transition has been observed in a well-known antiferromagnetic Mott insulator, kappa-ET_2 Cu[N(CN)$_2$]Cl[5]. As such sometimes frequency-dependent dielectric responses seem rather different from those observed in the quasi-one-dimensional TMTSF compound just above the charge order transition[6], its similarity with the glasses or other inhomogeneous systems had been discussed. Based on the quantum Monte Carlo simulation, an unbiased large-size numerical method, we now show that such kind of dielectric anomalies and ferroelectric-like transitions could be well explained in the context of conventional critical phenomena near the phase transition between the dimer Mott insulator and the charge order. The critical slowing down of the timescale and the extending correlation length scale between dimers localized on each dimers are analyzed. Such critical behavior of charges result in a peculiar nonuniform spatial structure of spin exchange interactions, which we call „spin animals". The spin animals contribute to the loss of coherent magnetic orders, which had previously been interpreted as an exotic spin liquid --- a lack of magnetism not like a paramagnet.

[1] M. Abdel-Jawad, et al, Phys. Rev. B 82, 125119 (2010).
[2] S.Iguchi, et al, Phys. Rev. B 87, 075107 (2013).
[3] M. Abdel-Jawad, et al, Phys. Rev. B 88, 075139 (2013).
[4] R. Okazaki, et al, JPS Conf. Proc. 3, 013010 (2014).
[5] P.Lunkenheimer, et al, Nat. Mater. 11, 755 (2012).
[6] P. Monceau, et al, Phys. Rev. Lett. 86, 4080 (2001).

14:15

Covalency-induced electric polarization in tetrathiafulvalene-p-chloranil (TTF-CA) studied by means of maximally-localized Wannier orbitals

S. Ishibashi[1,2], K. Terakura[1,3,4]
[1]*AIST, Nanomaterials Research Institute, Tsukuba, Japan;*
[2]*JST, CREST, Tokyo, Japan;*
[3]*National Institute for Materials Science, Tsukuba, Japan;*
[4]*Japan Advanced Institute of Science and Technology, Nomi, Japan*

The organic compound tetrathiafulvalene-p-chloranil (TTF-CA) forms a monoclinic crystal and, below ~81 K, its space group is non-centrosymmetric P1n1 [1]. Inherently, the system is a ferroelectric. Although theoretical calculations based on the Berry phase technique predicted large values of spontaneous polarization (8~10 μC/cm^2) [2], experimental confirmation was difficult because of leakage current. Recently, Kobayashi et al. successfully obtained a value of 6.3 μC/cm^2 and also found that the polarization vector is more than 20 times larger and opposite compared with that of the point charge model [3]. To clarify the origin of exotic ferroelectricity in TTF-CA, we made theoretical studies in terms of effective charges and maximally-localized Wannier orbitals (MLWOs) from first-principles calculations and compared with the case of $BaTiO_3$ [4]. Most recently, we have proposed a more explicit formulation to analyze the detailed mechanism of the covalency-induced polarization within the framework of MLWOs [5]. Application examples to TTF-CA as well as to perovskite transition metal oxides ($BaTiO_3$ and $PbTiO_3$) will be presented.

[1] M. Le Cointe et al., Phys. Rev. B 51, 3374 (1995).
[2] S. Ishibashi and K. Terakura, Physica B (Amsterdam) 405, S338 (2010); G. Giovannetti et al., Phys. Rev. Lett. 103, 266401 (2009).
[3] K. Kobayashi et al., Phys. Rev. Lett. 108, 237601 (2012).
[4] S. Ishibashi and K. Terakura, J. Phys. Soc. Jpn. 83, 073702 (2014); K. Terakura and S. Ishibashi, JPS Conf. Proc. 5, 011018 (2015).
[5] K. Terakura and S. Ishibashi, Phys. Rev. B, in press.

14:30

Current Switching Behaviour of Hydrogen-Bonding Ferroelectrics

T. Akutagawa[1,2], H. Anetai[2], T. Takeda[1,2]
[1]*Tohoku University, Institute of Multidisciplinary Research for Advanced Materials (IMRAM), Sendai, Japan;*
[2]*Tohoku University, Graduate School of Engineering, Sendai, Japan*

Organic materials with diverse molecular designs show multifunctional properties such as coupled ferroelectric, optical, ferromagnetic, and transport properties. We report the design of an alkylamide-substituted pyrene derivative displaying fluorescent ferroelectric properties coupled with electron transport properties. In solution phase, this compound displayed concentration-dependent fluorescence, whereas in xerogels, a fluorescent green organogel (>0.1 mM) and entangled nanofibers were observed. A discotic hexagonal columnar liquid crystalline phase was observed above 295 K due to intermolecular hydrogen bonding and p-stacking interactions. The direction of the hydrogen-bonded chains could be inverted by the application of an external electric field along the p-stacked column, resulting in ferroelectric polarisation-electric field (P-E) hysteresis. The local electric field arising from the ferroelectric macrodipole moment arrangement along the p-stacking direction affected the electron transport properties on the p-stack of pyrenes, thus confirming the current-switching phenomena according to P-E hysteresis (Fig.). We report that multifunctional properties such as ferroelectricity, fluorescence, and electron transport switching were successfully achieved in hydrogen-bonded dynamic p-molecular assemblies.

[1] Hayato Anetai, Yoshifumi Wada, Takashi Takeda, Norihisa Hoshino, Shunsuke Yamamoto, Masaya Mitsuishi, Taishi Takenobu, and Tomoyuki Akutagawa, J. Phys. Chem. Lett. 6, 1813–1818 (2015) 29 Yuta Shishido, Hayato Anetai, Takashi Takeda, Norihisa Hoshino, Shin-ichiro Noro, Takayoshi Nakamura, Tomoyuki Akutagawa*, J. Phys. Chem. C. 118, 21204-21214 (2014).*

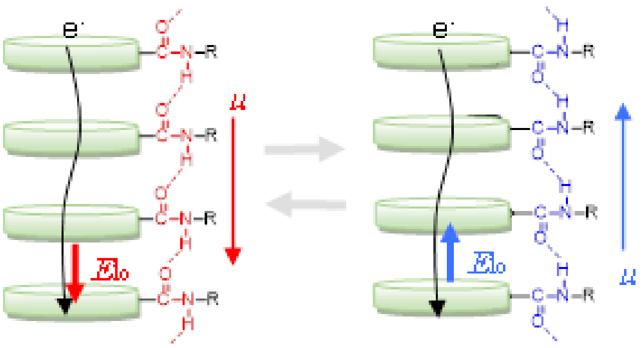


Figure: Carrier Transport Property in Col phase

Oral
Session 12

QUANTUM SPIN LIQUIDS II

Thursday, September 10, 2015

09:00 - 10:30

09:00

Proton-electron coupled functionalities based upon catechol-TTF based molecular materials

H. Mori
The University of Tokyo, The Institute for Solid State physics, Kashiwa, Japan

Hydrogen bonds in water, ice, DNA, proteins construct molecular assemblies by electrostatic interaction and play an important role in biological system. A proton in hydrogen-bond, light particle next to an electron, not only relates to construct solids, but also affords dynamical properties such as ferroelectrics and proton conductivity. However, these proton-related solid properties have been investigated, independent to electronic properties. In this research, proton-electron coupled purely organic functional materials have been developed [1] and characterized in order to explore novel electronic states concerted with proton dynamics. In this presentation, discoveries of (i) metallic state of purely organic single-unit materials under pressure [2], (ii) quantum spin liquid state of purely organic materials [3], (iii) switching of electrical and magnetic properties coupled to deuterium shift [4], (iv) structural and electronic transformation concerted with bending of hydrogen-bonded unit [5] will be introduced.

These researches are collaborated with Drs. A. Ueda, J. Yoshida (ISSP), A. Nakao (CROSS), R. Kumai, K. Kobayashi, H. Nakao, Y. Murakami (KEK), T. Isono, S. Uji (NIMS), K. Takahashi (Kobe Univ.), and Y. Nishio (Toho Univ.).

[1] H. Kamo, A. Ueda, H. Mori, et al., Tetrahedron Lett. 53, 4385-4388 (2012).
[2] T. Isono, A. Ueda, H. Mori, et al., Nature Commun. 4, 1344(1-6) (2013).
[3] T. Isono, A. Ueda, H. Mori, et al., Phys. Rev. Lett., 112, 177201(2014).
[4] A. Ueda, H. Mori, et al., J. Am. Chem. Soc., 2014, 136, 12184–12192. [5] J. Yoshida, A. Ueda, H. Mori,et al., Chem. Comuun., 2014, 55, 15557-15560.

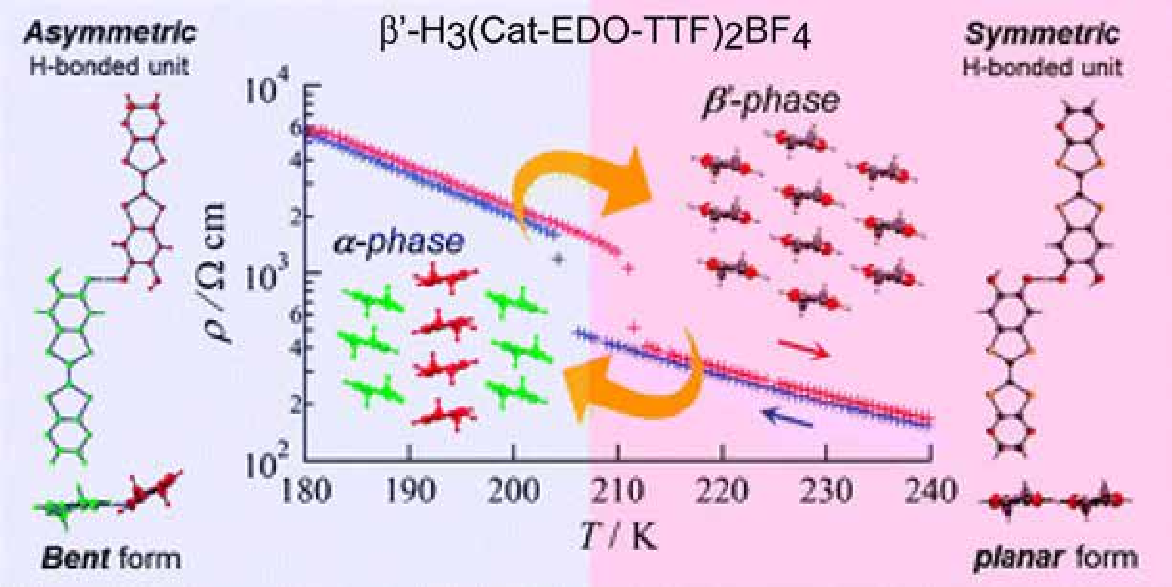


Figure 1: Phase transformation of novel proton-electron coupled molecular materials, beta'-H_3(Cat-EDO-TTF)$_2$$BF_4$.

09:30

Prediction of a spin-liquid ground state in the geometrically frustrated molecular crystal $Mo_3S_7(dmit)_3$

B. Powell, C. Janani, H. Nourse, A. C. Jacko, I. P. McCulloch, J. Merino
University of Queensland, Brisbane, Australia

We investigate quasi-one dimensional (q1d) models of $Mo_3S_7(dmit)_3$ based on triangles coupled into a chain. We discuss why a q1d electronic structure emerges from a 3d crystal structure [1]. We show, analytically, that when on-site Coulomb interactions spin-1 moments are formed on each molecule [2]. We then study this model using the density matrix renormalisation group and find that it gives rise to symmetry protected topological spin-liquid [3,4]. We show that this phase is adiabatically connected to the ground state of the spin-one Heisenberg model.

[1] A. C. Jacko, C. Janani, K. Koepernik, and B. J. Powell, Phys. Rev. B 91, 125140 (2015).
[2] C. Janani, J. Merino, I. P. McCulloch, and B. J. Powell, Phys. Rev. B 90, 035120 (2014).
[3] C. Janani, J. Merino, I. P. McCulloch, and B. J. Powell, Phys. Rev. Lett. 113, 267204 (2014).
[4] H. Nourse et al. unpublished.

10:00

Spatial symmetry breaking in κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ investigated by Terahertz Emission Spectroscopy

K. Itoh[1], H. Itoh[2,3], N. Yoneyama[4], T. Sasaki[1,3], S. Iwai[2,3]
[1]*IMR, Tohoku University, Sendai, Japan;*
[2]*Tohoku University, Sendai, Japan;*
[3]*JST-CREST, Sendai, Japan;*
[4]*University of Yamanashi, Kofu, Japan*

Charge order (CO) fluctuation in the organic dimer-Mott insulator is a subject of growing interest. In κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ (κ-CN), intra-dimer dipoles derived from CO were discussed[1,2], but the existence of dipoles and its collective phenomena is recognized as a controversial problem[3,4]. We performed Terahertz (THz) emission spectroscopy to observe the special symmetry breaking reflects spontaneous electric dipoles in κ-CN. Single crystalline κ-CN was irradiated with femtosecond infrared pulses (1.55 eV, ~25 fs), thus THz pulses were emitted by difference frequency generation if spatial symmetry is broken. THz detection was performed by typical THz time-domain spectroscopy system. Figure shows temperature dependence of emitted THz pulse intensity and inset shows emitted waveforms observed at 150 K and 30 K. THz emission is gradually available below 150 K and intensity increases with temperature reduces. This result indicates that spatial symmetry breaking is absolutely exist at low temperature. Considering the penetration depth of fundamental infrared pulses, emission efficiency at 30 K is about a half compared with a ZnTe crystal. This highly efficiency indicates that symmetry breaking could not be associated with extrinsic effects such as impurities and surface asymmetry. In addition, we observed polarity switching by applying positive or negative electric fields. It was noted that recent study[4] pointed out symmetry breaking due to CN disorder in the anion layer. Assuming their picture, replacement of atoms (C and N) would be needed to polarity inversion, but experiment fields (+-1 kV/cm) are too weak to cause such drastic modulation of atomic bonds. We will discuss more details compared with results in strict dimer-Mott insulator β'-(BEDT-TTF)$_2$ICl$_2$.

[1] Naka and Ishihara, J. Phys. Soc. Jpn. 82, 023701 (2013).
[2] Itoh et al., Phys. Rev. Lett. 110, 106401 (2013).
[3] Sedlmeier et al., Phys. Rev. B 86, 245103 (2012).
[4] Pinterić et al., Phys. Rev. B 90, 195139 (2014).

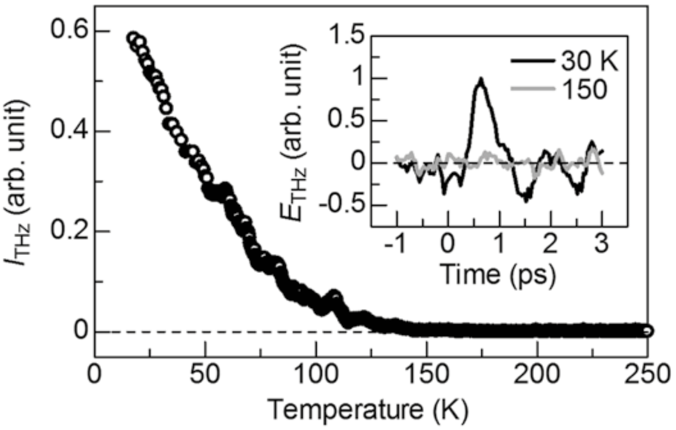


Figure: Temperature dependence of emitted THz pulse intensity and waveforms observed at 150 K and 30 K.

10:15

Spin correlation and dynamics of the spin-liquid material EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ revealed by in-plane ESR measurements

Y. Oshima[1], T. Tsumuraya[1,2], R. Kato[1]

[1]*RIKEN, Condensed Molecular Materials Lab., Wako, Japan;*

[2]*National Institute for Materials Science (NIMS), International Center forMaterials Nanoarchitectonics (WPI-MANA), Tsukuba, Japan*

The anion radical salts X[Pd(dmit)$_2$]$_2$, where X is a monovalent cation, are Mott insulators which consist of a S=1/2 triangular lattice. These salts have attracted attention over the years since the geometrical spin-frustration can be controlled by substituting the cation X. While most of them show a Néel order, the EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ shows no long-range order down to 30 mK. Although many studies suggest the title compound as a candidate material for the quantum spin-liquid state, the triangular lattice is strongly distorted and the nature of the ground state remains unclear. Therefore, the electronic states of EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ have been investigated by ESR measurements. As shown in the 'in-plane' angular dependence of the g-value (see Fig. 1), we have observed the splitting of two ESR absorptions coming from two independent anion layers in which the dimer stacking directions are different. This observation suggests the interlayer exchange interaction is infinitesimal, estimated ca. 0.37 mK, which is in a good agreement with the absence of long-range order in this salt. Moreover, the in-plane angular dependence of the ESR linewidth shows some features reflecting the non-equivalent triangular exchange interactions. Linewidth minima are observed when the magnetic field is applied along the strongest and second-strongest triangular exchange path, and a maximum is observed for the weakest triangular exchange path. Interestingly, the former ESR lineshape profile is a Lorentzian, while the latter is a Gaussian which might be due to the competition of two frustrating spin-chains. The characteristic ESR behavior of this salt will be presented in contrast with other salts that have different ground state, and its low temperature electronic state will be discussed.

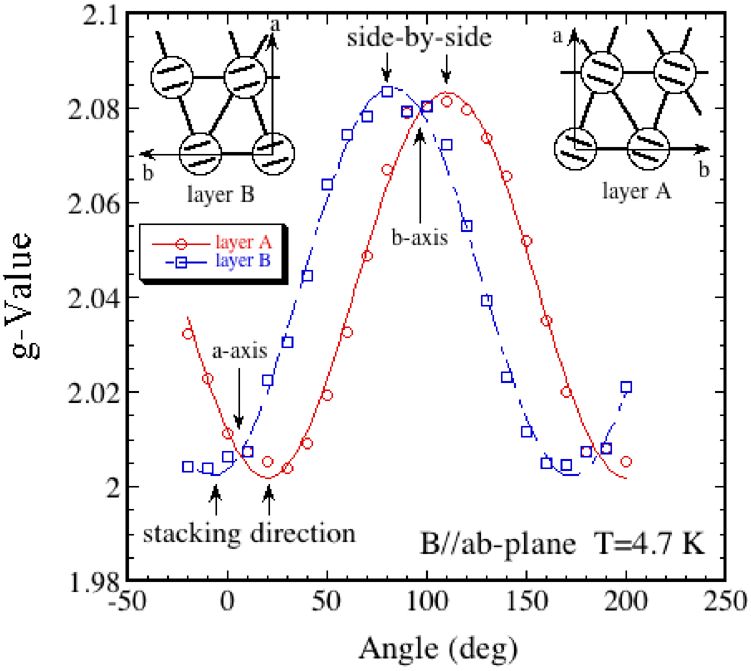


Figure 1: In-plane angular dependence of the g-value at 4.7 K

Oral
Session 13

FERROELECTRICITY II

Thursday, September 10, 2015

11:00 - 12:30

11:00

Donor-acceptor type organic ferroelectric crystals and thin films

S. Horiuchi[1,2], Y. Noda[1,2,] F. Kagawa[3,2,] K. Kobayashi[4], R. Kumai[4,2], T. Hasegawa[1]
[1]*National Institute of Advanced Industrial Science and Technology, Flexible Electronics Research Center, Tsukuba, Japan;*
[2]*Japan Science and Technology Agency, CREST, Tokyo, Japan;*
[3]*Riken, Center for Emergent Matter Science, Wako, Japan;*
[4]*High Energy Accelerator Research Organization, Institute of Materials Structure Science, Tsukuba, Japan*

Organic ferroelectric compounds which can exclude toxic lead and rare metallic elements are also promising for printed/flexible electronics, because of their light-weight, flexible, and soluble characteristics. Elaborate chemical designs are still required to meet the high performance, reliability, and process compatibility simultaneously. We have successfully improved the spontaneous polarization and Curie temperature so far on developing more than twenty examples of donor-acceptor (DA) type organic ferroelectrics in which the electric switching is based on the intra- and inter-molecular proton or electron transfer mechanisms. Among these, ferroelectric 2-methylbenzimidazole (MBI) has superior chemical stability, solubility, and polarization at room temperature [1] and then its crystalline thin-film can be fabricated by the solution process. The MBI crystal has switchable polarization with large out-of-plane component in the film, and allowed low-voltage switching as well as formation of locally inverted domains with piezoresponse force microscope (PFM). In this talk, I will report the development of DA-type organic ferroelectrics, switching process, and properties in bulk and thin-film forms.

[1] S. Horiuchi et al. Nat. Commun. 3:1308 (2012).

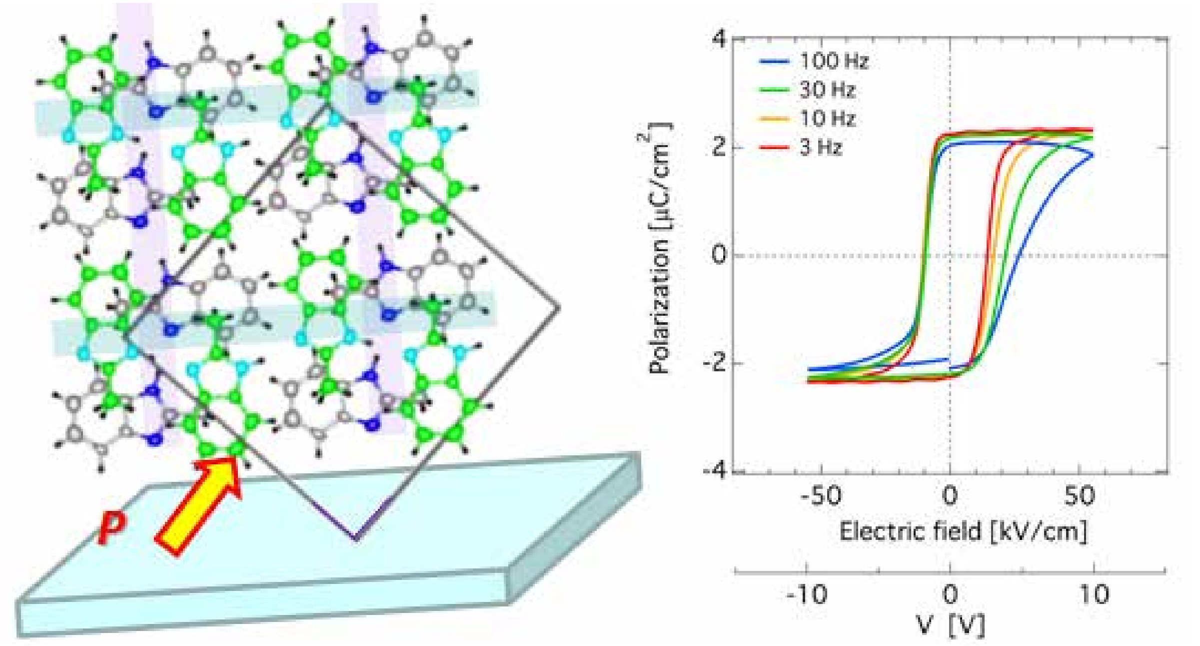


Figure 1: Molecular arrangegement and P-E hysteresis of MBI film.

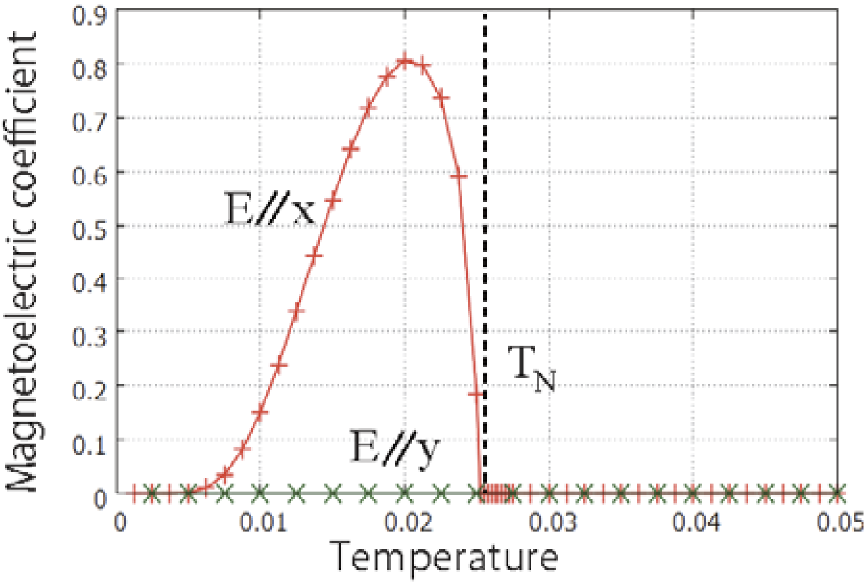
11:30

Novel dielectric, magnetic and magnetodielectric effects in dimer-type organic salts

S. Ishihara[1], M. Naka[1], J. Nasu[2], T. Watanabe[3]
[1]*Tohoku University, Department of Physics, Sendai, Japan;*
[2]*Tokyo Institute of Technology, Department of Physics, Japan;*
[3]*Chiba Institute of Technology, Department of Natural Science, Narashino, Japan*

Recently, molecular dimer-type low-dimensional organic compounds, such as kappa-(BEDT-TTF) $Cu_2(CN)_3$, $(TMTTF)_2AsF_6$, have much attention, since a number of exotic phenomena, i.e. spin-liquid, dielectric anomaly, superconductivity, have been observed. For a long time, electronic states in a series of compounds have tried to be understood in a single molecular orbital picture. Beyond this view point, we take the charge degree of freedom inside of the dimer units into account, and examine theoretically dielectric, magnetic and magneto-dielectric properties [1,2]. 1) We propose that the dimer Mott system provides a good stage for the linear ME effect [3]. We found that, in some magnetic and dielectric ordered states, the linear ME effect appears. This ME effect is also observed in spin- and charge-glass states due to a randomness. Essence of this effect is a hidden ferroic order of the spin and charge degrees of freedom inside of molecular dimer units. 2) We find a new type of the magnetic exchange interaction which favors a 90 degree angle between the neighboring spins [4]. This is a consequence of the spin-charge quantum entanglement inside of a dimer unit. A competition between this type interaction and the conventional Heisenberg interaction, we found that a magnetic moment for a conventional long-range mangetic order is strongly diminished by the quantum fluctuation. A possibility of the spin disordered state is suggested.

[1] S. Ishihara, J. Phys. Soc. Jpn. 79, 011010 (2010). (Review)
[2] S. Ishihara, J. Phys.: Cond. Matt. 26, 493201 (2014). (Review)
[3] M. Naka and S. Ishihara (in preparation)
[4] M. Naka, J. Nasu, T. Watanabe and S. Ishihara (in preparation).



Magnetoelectric effect: Temperature dependnce of magnetoelectric coefficient.

11:45

Electrodynamics and first-principles phonon calculations for the spin-liquid system κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

M. Dressel[1], P. Lazic[2], A. Pustogow[1], E. Zhukova[1,3,4], B. Gorshunov[1,3,4], J. A. Schlueter[5], O. Milat[6], **S. Tomić**[6]
[1]*Universität Stuttgart, 1. Physikalisches Institut, Germany;*
[2]*Rudjer Boskovic Institute, Zagreb, Croatia;*
[3]*RAS, A. M. Prokhorov General Physics Institute, Moscow, Russia;*
[4]*State University, Moscow Institute of Physics and Technology, Dolgoprudny, Russia;*
[5]*Argonne National Laboratory, Material Science Division, United States;*
[6]*Institut za fiziku, Zagreb, Croatia*

The dimer Mott insulator κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ showing an anomalous dielectric [1] and optical response [2] has been in the focus of intense activity in the last years. No definitive understanding has been achieved since experimental evidence [3] excludes theoretically predicted electric dipoles on molecular dimers [4]. Variable-range hopping in dc transport and dielectric response featuring relaxor-like ferroelectricity demonstrated an unforeseen inherent heterogeneity suggested to originate in the anion network [5]. Here we present broad-band electrodynamic investigations combined with first-principles density-functional calculations of the ground state and phonon vibrations [6]. The obtained results give insight into the origin of disorder within the anion network and the resulting domain structure. Moreover, a central role of the cation-anion coupling and the lattice phonons based on their joint movements in the anomalous terahertz response is revealed. We feel that the quantum spin liquid state in κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ can only be understood be taking the particular formation and coupling of the BEDT-TTF molecules to the Cu$_2$(CN)$_3$ anions into account.

[1] M. Abdel-Jawad et al., Phys.Rev. B 82, 125119 (2010).
[2] K. Itoh et al., Phys. Rev. Lett. 110, 106401 (2013).
[3] K. Sedlmeier et al., Phys. Rev. B 86, 245103 (2012).
[4] C. Hotta, Phys. Rev. B 82, 241104 (2010); M. Naka and S. Ishihara, J. Phys. Soc. Jpn. 79, 063707 (2010) and 82, 023701 (2013); H. Li et al., J. Phys.: Cond. Matt. 22, 272201 (2010); S. Dalay et al., Phys. Rev. B 83, 245106 (2011).
[5] M. Pinteric et al., Phys.Rev. B 90, 195139 (2014). [6] M. Dressel, to be submitted (2015).

12:00

Revisiting the charge-ordered α-(BEDT-TTF)$_2$I$_3$: Anisotropy of electronic properties

M. Čulo[1], M. Kuveždić[1,2], B. Mihaljević[2], E. Tafra[2], **M. Basletić**[2], T. Ivek[1], A. Hamzić[2], B. Korin-Hamzić[1], D. Schweitzer[3], M. Dressel[3], S. Tomić[1]
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α-(BEDT-TTF)$_2$I$_3$ is a quasi-2D organic conductor most famous for its prominent charge order within molecular planes below T_{CO} =136 K [1]. Due to the breaking of an inversion center, the response at the charge ordering transition should have a ferroelectric character [2,3]. Puzzlingly, at the moment it is unclear whether the ferroelectric peak appears in the dielectric properties at all [4]. Furthermore, the low-lying excitations above the ground state seem to bring forth both a phason-like as well as a soliton-like relaxation, which have so far not been reported to appear together in similar systems [5]. In order to explore these open questions, we have carefully determined the direction-dependent (in-plane and out-of-plane) dielectric response in α-(BEDT-TTF)$_2$I$_3$ together with resistance and Hall effect measurements down to lowest temperatures. The temperature dependence of the Hall coefficient absolute value follows a similar behavior as the resistivity. An intriguing feature is found at T_{CO} where the Hall coefficient changes sign. Another interesting property is a change in the in-plane transport activation energy around 60 K. On the other hand, dielectric measurements give strongly anisotropic and complex spectra. Our results are discussed within the wider context of strongly correlated systems with charge order.

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12:15

Dielectric spectroscopy on organic charge-transfer salts

P. Lunkenheimer[1], B. Hartmann[2], J. Fischer[1], M. Lang[2], J. Müller[2], S. Krohns[1], C. Mézière[3], P. Batail[3], A. Loidl[1]
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In this talk, we concentrate on the family of the two-dimensional BEDT-TTF-based charge-transfer salts, which in recent years have attracted considerable interest due to their often intriguing dielectric properties. For example, recently multiferroicity was found in κ-$(\text{BEDT-TTF})_2\text{Cu}[\text{N(CN)}_2]\text{Cl}$ (right frame of Fig. 1) and a new electric-dipole driven mechanism of multiferroicity was proposed to explain the unexpected fact that the magnetic and polar order appear at virtually identical temperatures in this system [1]. The polar moment in this system was proposed to be of predominantly electronic nature, arising from the dimerization of the BEDT-TTF molecules, combined with CO. A very recent example showing interesting dielectric behavior is α-$(\text{BEDT-TTF})_2\text{I}_3$, which will be treated in detail in this talk. In contrast to multiferroic κ-$(\text{BEDT-TTF})_2\text{Cu}[\text{N(CN)}_2]\text{Cl}$ and κ-$(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3$, the latter being a well-known relaxor system [2], CO in this material is well established. Recently relaxor-ferroelectric behavior was detected deep in the charge-ordered state of this system (left frame of Fig. 1) [3]. Again the dipolar degrees of freedom are assumed to be of electronic nature. Various dielectric investigations of this system exist and will be compared to the present results. Finally, we will present first results on δ-$(\text{EDT-TTF-CONMe}_2)_2\text{Br}$ [4], which exhibits CO already at room temperature but lacks any dimerization of the molecules. While we find evidence for two relaxational processes, there are no indications of polar order in this system.

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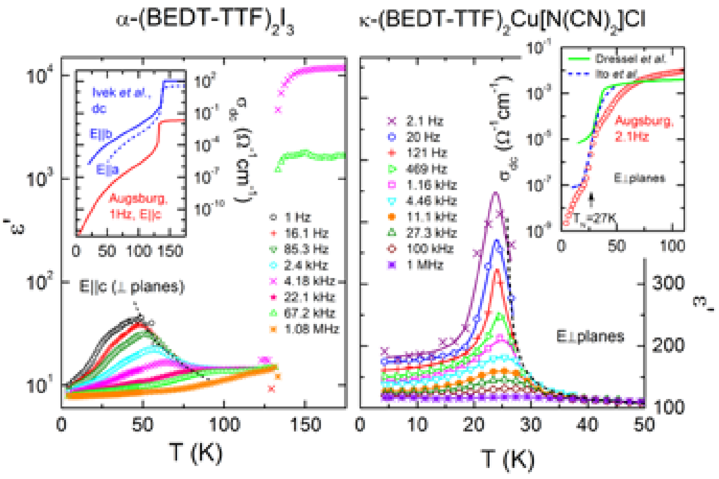

Figure 1: Temperature dependence of the dielectric constant of two two-dimensional charge-transfer salts, measured at various frequencies [1,3]. The insets show the dc conductivity taken from literature [5] and the present work

Oral
Session 14

DESIGN OF NEW MATERIALS II

Thursday, September 10, 2015

14:00 - 15:45

S14

14:00

Development of molecular conductors based on tetramethyl-TTP and its related donors

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In the development of molecular metals, a bis-fused TTF, 2,5-bis(1,3-dithiol-2-ylidene)-1,3,4,6-tetrathiapentalene (BDT-TTP) has played an important role, because BDT-TTP and its derivatives have produced a large number of stable metallic charge-transfer salts down to low temperatures (≤4.2 K).[1] Metallic state of most BDT-TTP conductors is significantly stabilized because of large bandwidth *(W)* and small on-site coulomb repulsion *(U)*. In this connection, control of to U/W is needed to realize superconductivity, that is, enhancement of *U* and/or reduction of *W*. Substitution with methyl groups might be effective to reduce bandwidth. We report herein synthesis of the tetramethyl derivative of BDT-TTP (TMTTP), TMTTP was prepared in 6% yield by $P(OMe)_3$-mediated cross-coupling reaction between 1,3-dithiole-2-thione fused with 4,5-dimethyl-TTF **(1)** and a large excess (30 eq. mol.) of 4,5-dimethyl-1,3-dihtiol-2-one **(2)** in refluxing toluene. X-Ray structure analysis of $(TMTTP)_2X$ ($X = ReO_4$, PF_6, AsF_6) revealed that the donors form two-dimensional conducting sheets (Figure 1). The packing pattern of the donors is classified as the so-called b-type (see also Figure 1). A tight-binding band calculation suggested that these salts have a quasi one-dimensional Fermi surfaces. All the salts showed high conductivity of $\sigma rt = 290 - 390\ S\ cm^{-1}$ on a single crystal. The PF_{6-} and AsF_{6-} salts exhibited metallic conductivity down to 10 K, while the ReO_{4-} salt showed metal to insulator transition at 126 K. Structures and conducting properties of molecular conductors based on the other alkyl substituted TTP derivatives will be also reported.

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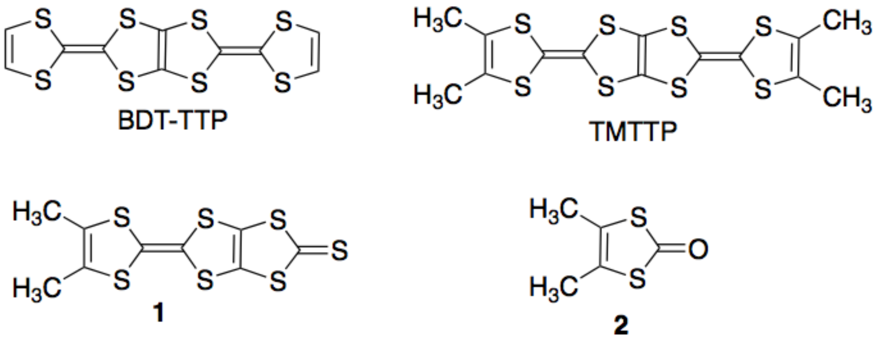

Chemical structures: Chemical structures of BDT-TTP, TMTTP, 1 and 2

14:30

Family of Q2D molecular conductors $(BEDT\text{-}TTF)_4(H_3O^+)[M^{III}(C_2O_4)_3]\times$Solvent: Mono- and bilayered phases, solvent-depending conductivity, phase transitions

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Past two decades the $(BEDT\text{-}TTF)_4(H_3O^+)[M^{III}(C_2O_4)_3]\times$Solvent family of molecular conductors remains one of the most enigmatic and exciting due to remarkable effect of guest solvent, included into hexagonal cavities of complex anion layer, onto crystal structure and properties. Some puzzles are very intriguing and still not fully solved such as mechanisms of switching on/off superconductivity by varying solvent or interlayer electron transport in bilayered phases which contain two BEDT-TTF layers of different packing and conductivity. The large title family involves four polymorphic phases of the same stoichiometry with different types of conducting BEDT-TTF layers including monolayered β", 'pseudo-κ' and bilayered α-β" and α 'pseudo-κ' [1-3]. Superconductivity is found only in the β"-salts of the family. The BEDT-TTF packing, formation of mono- or bilayered phases, symmetry of crystals as well as conductivity and temperature of superconducting transition depend on the size and symmetry of guest solvent molecule. Recently, using a modified procedure of the electrochemical synthesis, a large number of the β"-type crystals with different benzene and pyridine derivatives or mixtures of two solvents has been obtained. Detailed comparative analysis of their crystal structures reveals new aspect in the problem of solvent influence. Structural phase transition from monoclinic *C2/c* to triclinic *P*-1 symmetry observed at 180-230 K in several metallic and superconducting β"-salts with monohalogenated solvents is also discussed.

This work was supported by RFBR grant 14-03-00119 and Program of the Presidium of Russian Academy of Science.

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15:00

Bilayer molecular metals (CNB-EDT-TTF)$_4$X; A new prototype of 2D Molecular Conductors

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In this contribution a new type of 2D molecular conductors with a bilayer structure of the donors, based on the recently prepared dissymmetrical ET derivative cyanobenzene-ethylenedithio-tetrathiafulvalene (CNB-EDT-TTF) [1], is described. This donor through electrostatic and hydrogen bond assisted dimeric interaction involving the cyano groups promotes the formation of bilayers of the donors in salts of small anions X with composition (CNB-EDT-TTF)$_4$X. The characterization of salts with X= ClO_{4-}, PF_{6-}, and I_{3-}, by single crystal X-ray diffraction, electron transport measurements and band structure calculations will be presented showing how they present 2D metallic properties with unusual characteristics derived both from the unusual stoichiometry and the weak interaction between paired layers. As predicted by band structure calculations these properties are related with high band filling, large electronic effective masses and quasi degenerated 2D Fermi surfaces.

Work partially supported by FCT (Portugal) under contract PTDC/QEQ-SUP/1413/2012, UID/Multi/04349/2013 and doctoral grants of SO (SFRH/BD/72722/2010)

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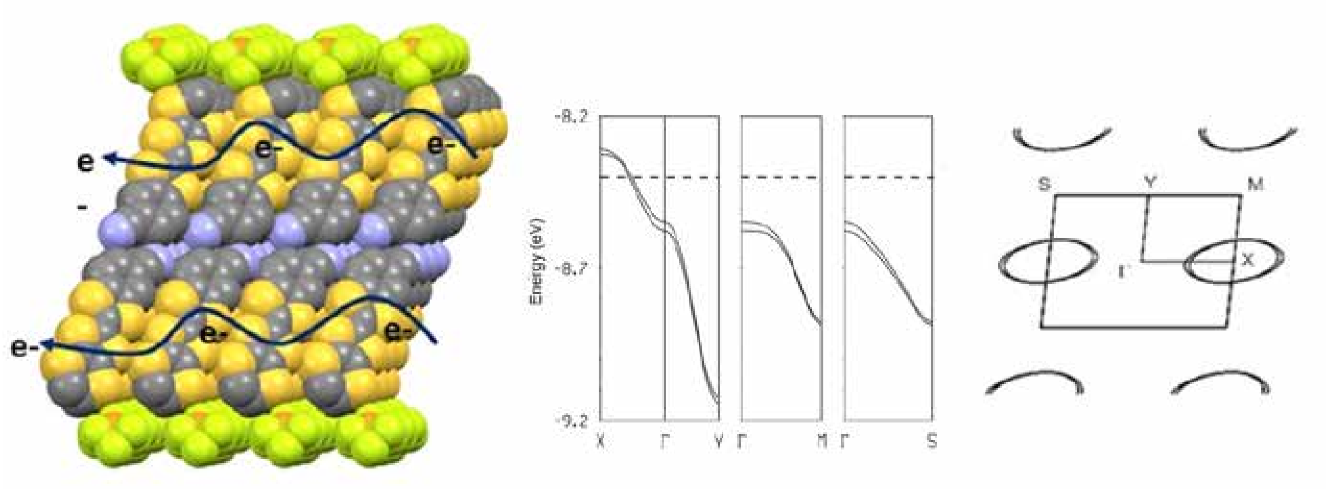

Bilayer conductor: Bilayer structure, calculated electronic band structure and Fermi Surface of (CNB-EDT-TTF)$_4$PF$_6$

15:15

Chiral conductors based on methylated TTF derivatives

F. Pop[1], G. Rikken[2], P. Auban-Senzier[3], E. Canadell[4], **N. Avarvari**[1]
[1] *CNRS-University of Angers, MOLTECH-Anjou, ANGERS, France;*
[2] *LNCMI, Toulouse, France;*
[3] *CNRS-University Paris Orsay, LPS, France;*
[4] *CSIC, ICMAB, Cerdanyola del Vallès, Spain*

Introduction of chirality into conducting systems is a topic of much current interest as it allows the preparation of multifunctional materials in which the chirality might modulate the structural disorder or expresses its influence through the electrical magneto-chiral anisotropy effect. The access to various chiral electroactive precursors for molecular conductors is therefore of paramount importance.[1] One of the strategies we have been developing in the last couple of years consists of the use of chiral methylated TTF (tetrathiafulvalene) derivatives such as TM-BEDT-TTF (TM = tetramethyl, BEDT = bis(ethylenedithio), DM-BEDT-TTF (DM = dimethyl), and DM-EDT-TTF (EDT = ethylenedithio) in electrocrystallization experiments in the presence of various anions.[2,3] For example, the radical cation salts of DMEDT-TTF and PF_{6-} show semiconducting properties in the enantiopure form and metallic conductivity for the racemic form.[4] Investigations of the structural features and conducting properties of complete series of radical cation salts (enantiopure and racemic forms) from these families of derivatives will be presented. A special attention will be given to the first experimental observation of the electrical magneto-chiral anisotropy effect in a bulk chiral conductor.[5]

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15:30

Molecular alloys of neutral gold/nickel dithiolene complexes in single component semiconductors

M. Fourmigué[1], K. Mebrouk[1], W. Kaddour[2], P. Auban-Senzier[2], C. Pasquier[2], O. Jeannin[1], F. Camerel[1]
[1]*Université Rennes 1 & CNRS, ISCR, France;*
[2]*Université Paris Sud, LPS, Orsay, France*

Control of band filling or doping of molecular (semi)conductors can be performed by substitutional insertion of molecules with similar shape but a different electron count, with one more or one less electron [1]. This strategy has been explored here within the semiconducting, single-component, radical gold dithiolene complex [$AuOC_4$] bearing para-butoxyphenyl substituents [2]. Alloying with the corresponding neutral nickel dithiolene complex [$NiOC_4$] lacking one electron afforded a complete isostructural series $[NiOC_4]_{1-x}[AuOC_4]_x$, spanning the whole composition range from x = 0 to x = 1 by 0.1 increments, further characterized by X-ray diffraction and EDX analyses. Magnetic susceptibility data confirm the antiferromagnetic interactions between neighboring radical gold dithiolene complexes. The electrical conductivity increases exponentially with the x gold fraction, while the activation energy remains constant in the more conducting, gold-rich samples. This behavior is tentatively assigned to the tunneling barriers of variable width (with x) but of constant height, separating more conducting gold-rich segments [3]. Comparison of redox potentials for the 1e– oxidation and reduction of both [$NiOC_4$] and [$AuOC_4$] dithiolene complexes indicates that the [$NiOC_4$] nickel complex does not act as a dopant for the radical [$AuOC_4$] complex [4].

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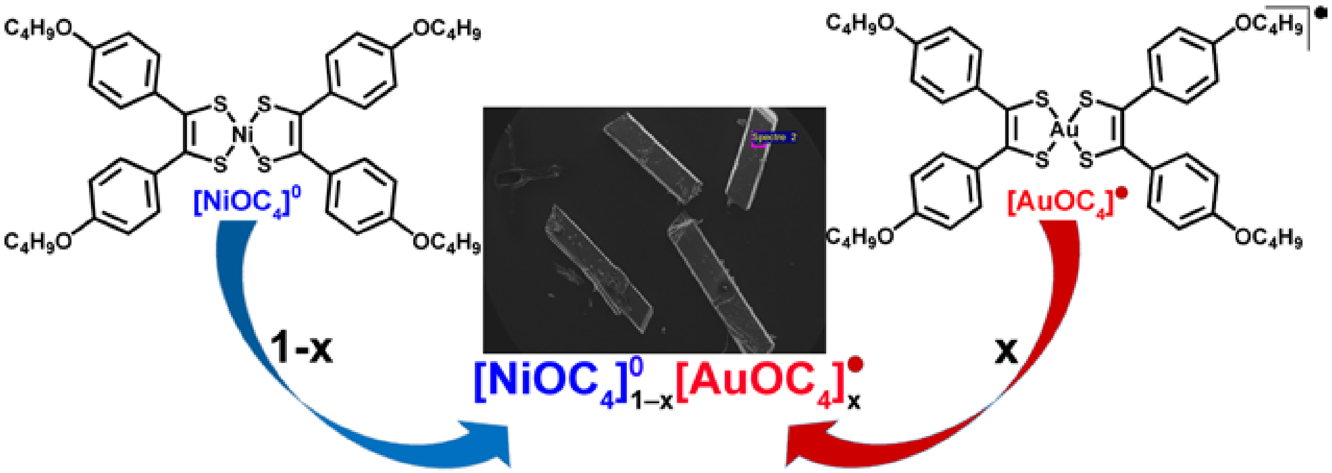

Figure 1: Molecular alloys of neutral gold/nickel dithiolene complexes

Oral
Session 15

MOTT PHYSICS II

Thursday, September 10, 2015

16:15 - 17:30

16:15

Randomness effect on the correlated electrons in the molecular conductors irradiated by X-ray

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Mott-Anderson transition has been known as a metal-insulator (MI) transition caused by both strong electron-electron interactions and randomness potential modulation for the electrons. For example, the MI transition in doped semiconductors and transition metal oxides has been investigated as a typical example of the Mott-Anderson transition for changing electron correlations by carrier number control in concurrence with inevitable randomness. On the other hand, molecular conductors have been known as typical strongly correlated electron systems with bandwidth controlled Mott transition. In this talk, our recent progress of the research on the randomness effect on the Mott transition in k-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Y system is presented. X-ray irradiation on the crystals introduces molecular defects of -CN part in the insulating anion layer, which cause random potential modulation for the correlated electrons in the conductive BEDT-TTF layer. In combination with hydrostatic pressure and chemical substitution, we are able to control the parameters for randomness and correlations in pi-electrons approaching the Mott-Anderson transition.

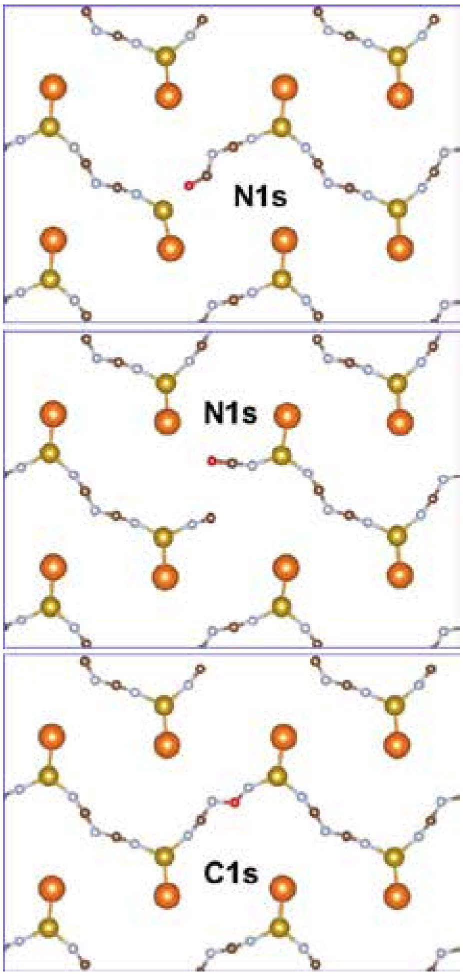

Figure 1: Optimization of anion network structure introducing a core hole in C1s and N1s.

16:45

Detailed study of the Mott transition in $EtMe_3P[Pd(dmit)_2]$

M. Abdel Jawad[1], R. Kato[1], I. Watanabe[2], N. Tajima[3], Y. Ishii[4]
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[2]*RIKEN-RAL, Nishina Centre, Wako, Japan;*
[3]*Toho University, Department of Physics, Funabashi, Japan;*
[4]*Shibaura Institute of Technology, Department of Physics, College of Engineering, Saitama, Japan*

Many questions related to the metal-insulator transition of the Mott type remains unanswered to this day. In this presentation, we will present isothermal pressure measurements of the conductivity, σ, the thermoelectric effect and the Hall coefficient within a Helium pressure medium in a dimer-Mott insulator with the valence bond order state, $EtMe_3P[Pd(dmit)_2]_2$. One surprising result is that the Seebeck coefficient, SB can determine the critical pressures of the Mott transition in a clear and objective way [1] (see figure). However, this estimate of the Mott transition contradicts with the assumptions of previous studies [2,3]. In another surprising result, which is related to the Hall coefficient measurements, the carrier number of $EtMe_3P[Pd(dmit)_2]_2$ is found to be almost temperature and pressure independent in its metallic state. Discussions of the universality class of the Mott transition will be made and compared to previous results in other Mott insulators [2,3,4].

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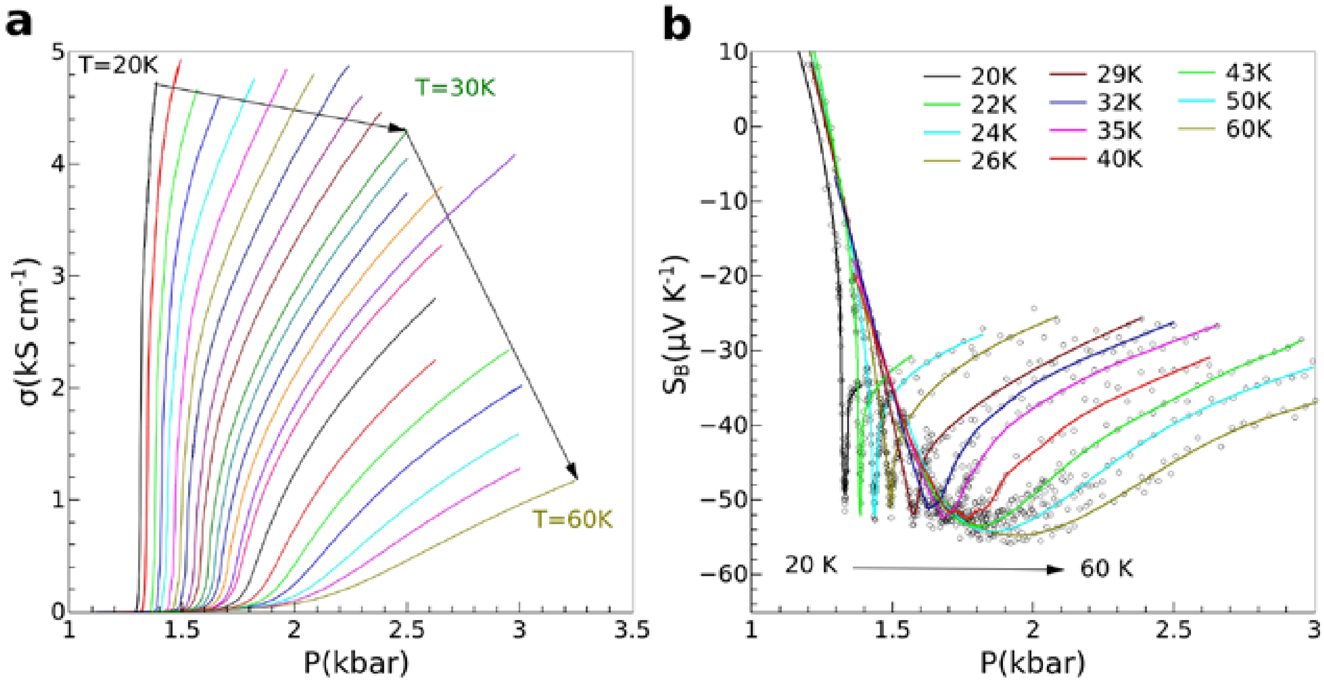

Figure: Isothermal pressure dependence of the conductivity (a) and the Seebeck coefficient (b) in $EtMe_3P[Pd(dmit)_2]_2$ at temperatures ranging from 20 K up to 60 K.

17:00

Antiferromagnetic fluctuations in organic superconductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br detected by Raman spectroscopy.

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[3]*Argonne National Laboratory, Materials Science Division, United States;*
[4]*The National Science Foundation, Division of Materials Research, Arlington, United States*

We present magnetic Raman spectra of a quasi-two dimensional organic superconductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br at temperatures above the superconducting transition. We show that on cooling below 200 K in the in-plane cross-polarization (B_{2g} symmetry) the spectra develop a broad band at about 500 cm^{-1} which we associate with two-magnon excitations. Both the position and the temperature dependence of the spectral weight of the two-magnon excitations in the Fermi-liquid state of κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br are similar to that of an antiferromagnetic Mott insulator κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl. We conclude that the strength of two-dimensional anti-ferromagnetic correlations, which define the two-magnon in-plane response, is the same in antiferromagnetic insulating and Fermil-liquid metallic states. The fact that metallic properties in κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br are obtained by reducing electronic correlations compared to the antiferromagnetically ordered Mott insulator state while preserving stoichiometry can account for a strength and position of the two-magnon band.

17:15

Gold dithiolene complexes as Mott Insulators

D. Lorcy[1], Y. Le Gal[1], A. Filatre-Furcate[1], N. Bellec[1], P. Auban-Senzier[2]
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[2]*Université de Paris-Sud, Laboratoire de Physique des solides, Orsay, France*

Beyond the well-developed use of metal bis(dithiolene) complexes as precursors of multi component molecular conductors in mixed-valence salts, an emerging trend is to use these complexes as precursors of single component molecular conductors [1]. We recently prepared a series of single component molecular conductors based on neutral radical gold dithiolene complexes derived from [Au(Et-thiazdt)$_2$] which are pressure sensitive [2]. These single component molecular conductors behave as Mott insulator with one radical per site at variance with organic conductors with mixed valence character. The greater sensitivity of these neutral single component conductors to external stimuli, as compared with „classical" radical salts, makes these compounds valuable candidates for the use of other perturbation than pressure such as electric pulses of interest for information storage [3]. Herein, we wish to present a series of single-component conductors derived from [Au(R-thiazdt)$_2$] where we tested the influence of the R group on the organization and the conducting properties of the neutral radicals [Au(R-thiazdt)$_2$]$^{\bullet}$.

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Oral
Session 16

ULTRAFAST PHENOMENA

Friday, September 11, 2015

09:00 - 10:30

09:00

Strong field effects on organic conductors induced by nearly single-cycle light pulse

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Ultrafast control of conduction and magnetic properties in strongly correlated systems has been extensively studied from the perspective of photoinduced insulator-to-metal transitions, or equivalently, the “melting” of electronic orders in Mott insulators, charge ordered systems. On the other hand, the development of strong electric fields (> MV/cm) of few-cycle optical pulses and recent theoretical studies using dynamical mean-field theory (DMFT) [1] suggest that extreme non-equilibrium electronic states such as cahrge localization, negative temperatures, and repulsive-attractive conversion can be achieved. Among those highly non-equilibrium phenomena, reducing the intersite transfer integral t by modulating the site energy under shigh-frequency [ω>t/(h_bar)] continuous-wave (CW) and pulsed AC electric fields E(ω) have attracted attention for > 30 years [2,3]. Such an intense modulation of the electronic structure driven by a strong electric field, referred to as „dynamical localization“, provides a new strategy for controlling charge motion in strongly correlated materials, which have a competing energy balance between the on-site or intersite Coulomb repulsion and t. Here we describe a charge localization induced by the 9.3 MV/cm instantaneous electric field of a 1.5 cycle (7 fs) infrared pulse in an organic conductors α-$(ET)_2I_3$ (ET; bis[ethylenedithio]-tetra-thiafulvalene)[4], $(TMTTF)_2AsF_6$ (TMTTF; tetramethyltetrathiafulvalene). A large reflectivity change of > 25% and a coherent charge oscillation along the time axis reflect the opening of the charge ordering gap in the metallic phase. This optical freezing of charges, which is the reverse of the photoinduced melting of electronic orders, is attributed to the 10% reduction of t driven by the strong, high-frequency electric field.

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09:30

Control of electronic interactions in organic conductors and superconductors

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The interplay of correlations and structural dynamics that leads to electronic order in the Mott transition or Charge Density Wave states is one of the key parameters in correlated electron systems. It gives rise to the large variety of different ground states ranging from bad metallic states to superconductivity. Here we set out to control these electronic interactions using high intensity light pulses establishing new dynamic states in the material. In order to understand basic concepts of dynamically driven states in complex correlated systems we want to discuss the dynamics in the organic charge transfer salt ET-F_2TCNQ, a prototypical 1D Mott insulator. NIR light pulses excite charge carriers above the Mott gap to induce a photometallic state. The optical response of the charge transfer reveals an ultrafast dynamics of a doublon-holon system controlled by the nearest neighbor interactions V [1], tunable by external pressure [2]. In contrast to that exciting the system resonant at specific molecular vibrations of the ET molecule we achieve control of the molecular electronic wavefunction. In that way we modulate the on-site two particle Coulomb repulsion and Mott criterion U/t and observe the properties of the system in the dynamically driven state [3,4]. These open novel ways of controlling effective interactions and functionalizing complex correlated materials. As an example we show how we can use these to transiently promote superconductivity above T_c in the organic superconductor K_3C_{60}.

[1] S. Wall et al. Nat. Phys. 7, 114 (2011).
[2] M. Mitrano et al. Phys. Rev. Lett. 112, 117801 (2014).
[3] S. Kaiser et al. Scientific Reports 4, 3823 (2014).
[4] R. Singla et al. arxiv:1409.1088 (2014).

10:00

Coherent excitations at the neutral-ionic transition: Femtosecond dynamics on diabatic potential energy surfaces

A. Painelli[1], L. Cavatorta[1], Z. Soos[2]
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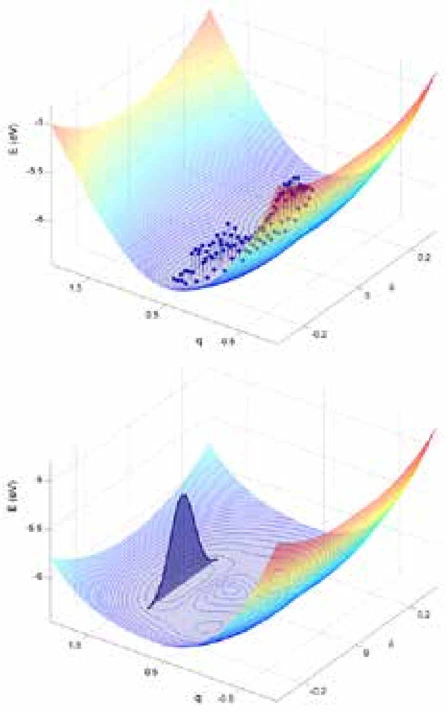
Mixed-stack charge-transfer (MS-CT) crystals offer an interesting opportunity to study strongly correlated electrons coupled to molecular vibrations and lattice modes. The most impressive demonstration of strong correlations in these systems is the neutral-ionic phase transition (NIT), a transition from a band insulator to a Mott insulator that can be induced by temperature, pressure, or light. NIT is always accompanied by lattice dimerization, eventually leading to ferroelectric behaviour. Okamoto et al. [1] recently explored the early-stage dynamics (0-3 ps) of the photoinduced NIT in TTF-CA, via ultrafast pump-probe measurements. The generation of ionized domains through the absorption of a single photon (multiexciton generation) was observed along with the modulation of the frequency of molecular modes by the lattice mode. The minimal description of NIT is offered by a modified Hubbard model with linear coupling to a molecular vibration and to the Peierls phonon. The model was successfully applied to ground state properties of MS-CT, including the charge distribution and lattice structure, vibrational spectra, diffuse X-ray scattering data, and the dielectric anomaly [2]. Here apply the same model to the early time evolution of a photoexcited system. Complex adiabatic potential energy surfaces (PES) signal multistability. To address the photoinduced dynamics we define diabatic PES, corresponding to the different stable and metastable states of the system. We reproduce coherent oscillations of molecular vibrations and of the Peierls mode following ultrafast excitation and quite naturally explain the modulation of vibrational frequencies by the Peierls mode in terms of strong anhamonicity. [3]

[1] H. Uemura, H. Okamoto, Phys. Rev. Lett. 105 (2010) 258302.
[2] G. D'Avino, et al., Phys. Rev. B 83 (2011) 161105.
[3] L. Cavatorta, A. Painelli, Z. G. Soos, Phys. Rev. B 91 (2011) 174301.



Ground and metastable PES

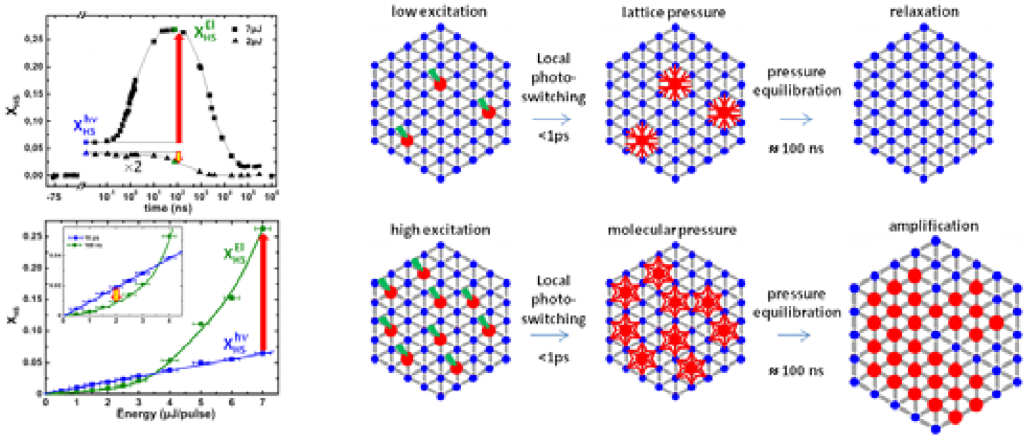
10:15

Real-time cooperative response to light in breathing crystals driven by a photoinduced elastic field

E. Collet, R. Bertoni, M. Cammarata, H. Cailleau, M. Lorenc
University Rennes 1, Institut de physique de rennes, France

Ultrafast photoswitching in bistable spin-crossover molecular crystals is associated with a complex transformation pathway, multiscale in nature, where both molecular photo-switching and macroscopic transformation of the crystal have to be considered [1-3]. We have studied the basic mechanisms allowing light to switch the molecular state, from low spin (LS) to high spin (HS). Femtosecond x-ray absorption (LCLS X-FEL) and optical spectroscopy show that the stabilization of the photoinduced HS state results from self-trapping of the electronic excited state. The stabilization of the newly formed electronic state results from a two steps relaxation sequentially involving molecular stretching and torsion accompanied by the generation of coherent molecular phonons. The system is bobsledding across a potential energy surface along identified coordinates [4-5] In the active medium, which the crystal is, other effects of elastic (propagating) or thermal (diffusive) nature should be considered [1-3]. In the case of cooperative solids, a self-amplified and coherent response to light excitation is observed on short time-scales. When a certain fraction of molecules, initially in LS state, is photo-converted to HS state (of higher molecular „volume") within 200 fs, the resulting internal pressure drives lattice expansion. This self-amplification process results from the elastic field induced by light and coupled to the molecular volume change. This illustrates the efficiency of cooperative mechano-elastic process during photo-induced transformation in a material. Our study brings the photo-switching of materials into new perspective, notwithstanding its common perception, uniquely related to electronic or optical and acoustic phonon dynamics.

[1] M. Lorenc et al, Phys. Rev. Lett., 103, (2009) 028301.
[2] R. Bertoni et al, Coord. Chem Rev. 282-283, (2015) 66-76
[3] E. Collet et al Physical Review Letters, 109, (2012) 257206
[4] M. Cammarata et al, Physical Review Letters, 113, (2014) 227402
[5] R. Bertoni et al, Account Chem. Res. 48, (2015) 774-781



Cooperative response: a) Time evolution of the HS fraction for low and high excitation. b) dependence of the HS fraction after photoexcitation (10 ps) and on the elastic step (100 ns). Schematic representation.

Oral
Session 17

FUNCTIONAL MATERIALS - INTRAMOLECULAR PROPERTIES

Friday, September 11, 2015

11:00 - 12:00

11:00

Using muons to probe order and excitations in molecule-based spin chains and ladders

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[2]*Oxford University, Physics, Great Britain;*
[3]*ISIS Facility, Muon group, Didcot, Great Britain;*
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One-dimensional quantum magnetism continues to be of great theoretical and experimental interest, as reduced dimensionality supports strong quantum fluctuations which can result in novel excitations and critical behaviour. In recent years, coordination polymers have provided materials that are excellent realisations of low-dimensional theoretical models, with an added potential for tuning interaction strengths by varying the molecular building blocks comprising the materials. Here we discuss our recent results in this area, with an emphasis on the use of muon-spin relaxation as a sensitive probe of emergent magnetism in spin-1/2 chains and ladders. Muons have repeatedly been shown to be sensitive to long-range magnetic order in these systems [1,2], which is often very difficult to observe using other techniques. In addition, muons are also very sensitive probes of low-energy dynamics. We will present results where we have observed spin diffusion in the highly ideal one-dimensional antiferromagnetic spin-chain $Cu(pyz)(NO_3)_2$ [3], along with investigations of the different dynamical character of molecular spin ladders in the strong-rung and strong-leg regimes. These measurements address long-standing questions about the nature of excitations in one-dimensional systems and demonstrate several state of the art uses of muon spectroscopy.

[1] T. Lancaster et al., Phys. Rev. B 73, 020410 (2006).
[2] T. Lancaster et al., Phys. Rev. Lett. 112, 207201 (2014).
[3] F. Xiao et al., Phys. Rev. B 91, 144417 (2015).

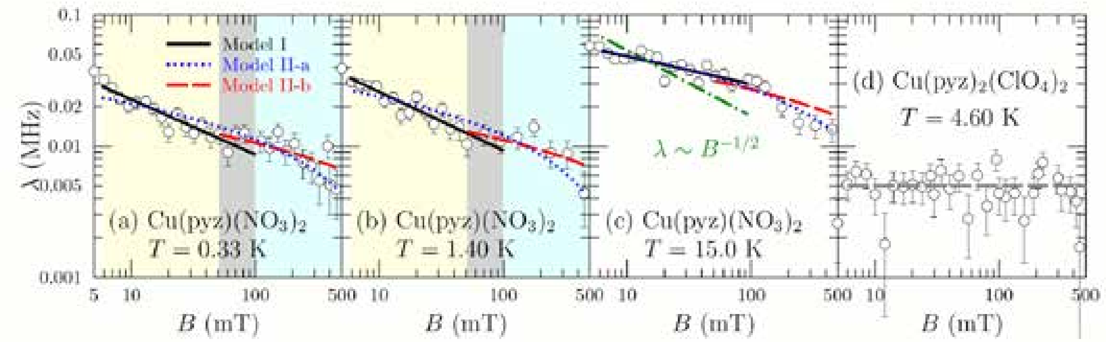

Figure 1: Muon-spin relaxation rates in one-dimensional $Cu(pyz)(NO_3)_2$ [3], allowing the identification of diffusive spin dynamics.

11:30

Mixed stack CT crystals: old materials still holding the scene

A. Girlando
Parma University, Chemistry, Italy

Since their discovery mixed stack (ms) CT crystals have alternated periods of great interest to periods of almost complete oblivion. During the quest for organic metals, all synthesized ms crystals very discarded as uninteresting semiconductors or insulators. Attention resurrected with the discovery of neutral-ionic transition, with associated exotic phenomena [1], including photoinduced transitions [2]. Ionic ms crystal may become ferroelectric [3], or even multiferroic [4]. More recently, ms CT crystals have been proposed as ambipolar semiconductors [5]. In this talk I will present several new examples of ms CT crystals. A new series based on acenes and fluorine substituted TCNQ allow to investigate the effect of the degree of ionicity and of different stoichiometries on their physical properties. Two old donors, 3,3‘,5,5‘-tetramethyl-benzidine and 1,1,4,4-tetrathio-butadiene, associated with both TCNQ and haloquinone acceptors, allow one to span the whole range of ionicities and to discover new systems undergoing the neutral-ionic transition. A unified picture of the varied and unusual properties of ms crystals can only be obtained by keeping in mind that they are highly correlated systems, and on this basis a sort of phase diagram is proposed.

[1] J.B. Torrance et al., Phys. Rev. Lett. 47, 1747 (1981); Z.G. Soos et al. , Synth. Metals 155, 357 (2005).
[2] M. Gonokami and S. Koshihara, J. phys. Soc. Japan 75, 0110011 (2006).
[3] S. Horiuchi et al., Chem. Lett. 43, 26 (2014).
[4] G. Giovannetti et al., Phys. Rev. Lett., 103, 266401 (2009).
[5] L. Zhu et al., J. Am. Chem. Soc. 134, 2340 (2012).

11:45

Stochastic magnetization jumps in chiral molecular magnets

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Molecular magnets of chiral magnetic structure attract great attention due to new physical phenomena found in the last years: non linear spin excitations, bistability of ferromagnetic resonance, and domain walls damping in periodical Peierls relief. In the recent theoretical work [1], jumps of magnetization in an incommensurate spin-soliton lattice were predicted. Our purposes are experimental observing and identification of magnetization jumps in $[Mn((R/S)\text{-}pn)]_2[Mn((R/S)\text{-}pn)_2(H_2O)][Cr(CN)_6]_2$ chiral magnets described in [2,3]. We used SQUID magnetometer to obtain magnetic hysteresis loops of the sample at 2-50 K. The static magnetic field was stepwise swiped with scanning step 0.2-2 Oe in the 0-50 kOe range. Series of the magnetization jumps was observed. Despite good reproducibility of the magnetization jumps, individual critical magnetic fields as well as amplitude of each jump were casual, i.e. magnetization jumps were stochastic. The starting field of magnetic jumps decreases as sample was heated. Observed jumps caused by quantized energy of spin soliton lattices of different periods. Transitions between these states was observed as steps on the field dependence of the magnetization. Temperature dependencies of critical magnetic field initiating demagnetization jumps are correlation with magnetic anisotropy constant. Thus magnetization jumps caused by spin soliton formations were experimental observed in $[Mn((R/S)\text{-}pn)]_2[Mn((R/S)\text{-}pn)_2(H_2O)][Cr(CN)_6]_2$ molecular magnets.

The work was supported by the RFBR grant 15-02-05149.

[1] J. Kishine, G. Bostrem, A.S. Ovchinnikov et al., Phys. Rev. B 89, 014419 (2014).
[2] J. Kishine, K. Inoue, Y. Yoshida, Prog. Theor. Phys. Suppl. 159, 82 (2005).
[3] R.B. Morgunov, M.V. Kirman, K. Inoue et al., Phys. Rev. B 77, 184 419 (2008).

Poster Session I

Tuesday, September 8, 2015

20:00 - 22:00

Metal-Insulator transition of the Field Induced Superconductor, λ-BETS$_2$FeCl$_4$; Studied by microscopic point of view

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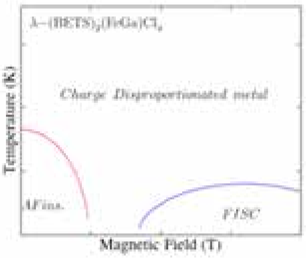
[1]*Gakushuin University, Physics, Tokyo, Japan;*
[2]*Institute for Molecular Science, Okazaki, Japan;*
[3]*Nihon University, Department of Chemistry, Tokyo, Japan;*
[4]*RIKEN, Wako, Japan*

The molecular based charge transfer complex, λ-(BETS)$_2$FeCl$_4$, which is known as a field induced superconductor [1]. The field induced superconductivity was caused by the so-called ``compensation" mechanism as experimentally confirmed by several experimental studies including our ^{77}Se NMR study at high magnetic field [2]. On the other hand, the system exhibits a metal-insulator transition with magnetically ordering at lower field region [3]. The schematic temperature-field phase diagram is shown in the Figure 1. The antiferromagnetic insulating state has been believed as a cooperation between conduction π spins on organic BETS molecule and localized d spins on Fe^{3+} site. On the other hand, an idea which explains the antiferromagnetically ordering (AFO) insulating state has been proposed by the analysis of the specific heat measurement; p spins contribute to the AFO state but the localized d spins remain paramagnetically [4]. To detect p spin dynamics on the organic BETS site, temperature/field dependences of ^{77}Se and ^{13}C NMR measurements using a single crystal sample were performed. Relatively narrower ^{77}Se/^{13}C-NMR spectra were observed at high temperature metallic state. The spectral width became broader and 1/(T1T) increased with cooling. At low temperatures, the system exhibits AF-LRO at ~7 K/3K under 4T/9T. This strongly suggests the existence of an antiferromagnetic interaction/fluctuation between conducting π spins. The spin dynamics at low magnetic field region of the system will be discussed

[1] L. Balicas et al., Phys. Rev. Lett. 87, 067002 (2001)
[2] K. Hiraki et al., J. Phys. Soc. Jpn. 76, 124708 (2007)
[3] S. Uji et al., J. Phys. Soc. Jpn. 72, 369 (2003)
[4] H. Akiba et al., J. Phys. Soc. Jpn. 78, 033601 (2009)



Schematic phase diagram.: phase diagram

PI-2

Anomalous charge response in a novel spin-liquid candidate κ-$(BEDT\text{-}TTF)_2Ag_2(CN)_3$

M. Kuveždić[1,2], M. Pinterić[1,3], **T. Ivek**[1], M. Basletić[2], B. Korin-Hamzić[1], O. Milat[1], T. Hiramatsu[4], G. Saito[4], S. Tomić[1]
[1]*Institut za fiziku, Zagreb, Croatia;*
[2]*University of Zagreb, Faculty of Science, Department of Physics, Croatia;*
[3]*University of Maribor, Faculty of Civil Engineering, Slovenia;*
[4]*Meijo University, Faculty of Agriculture, Nagoya, Japan*

Novel forms of the low-temperature phases in the two-dimensional molecular solids with competing interactions between charges, spins and lattice, in particular those featuring anomalous dielectric relaxation, have been the focus of intense activity in recent years. Open issues concern the nature of collective charge excitations as well as their coupling to applied ac and dc electric fields. Dimer Mott insulator with quantum spin-liquid phase κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$ shows variable-range hopping in dc limit and dielectric response featuring relaxor-like ferroelectricity [1,2] despite missing symmetry-breaking evidences [3]. In this presentation we report our study of the anisotropic dc and ac charge response in a novel spin-liquid candidate κ-$(BEDT\text{-}TTF)_2Ag_2(CN)_3$ [4]. The obtained results indicate the presence of an anomalous dielectric response, and we discuss and compare them with κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$. The importance of anion network geometry and cation-anion coupling is emphasized.

[1] M. Abdel-Jawad et al., Phys.Rev.B 82, 125119 (2010).
[2] M. Pinterić et al., Phys.Rev.B 90, 195139 (2014).
[3] K. Sedlmeier et al., Phys.Rev.B 86, 245103 (2012).
[4] M. Pinterić et al., in preparation (2015).

PI-3

Emergence of the Jahn–Teller metallic state in alkali metal fulledies shown by infrared spectroscopy

G. Klupp[1], P. Matus[2], R. H. Zadik[1], R. H. Colman[1], Y. Takabayashi[1], M. J. Rosseinsky[3], K. Kamarás[2], K. Prassides[4]
[1]*Durham University, Department of Chemistry, Great Britain;*
[2]*Wigner RCP, Institute for Solid State Physics and Optics, Budapest, Hungary;*
[3]*University of Liverpool, Department of Chemistry, Great Britain;*
[4]*Tohoku University, World Premier International–Advanced Institute for Materials Research, Sendai, Japan*

The unconventional superconducting phase of A_3C_{60} fullerides emerges from the Mott–Jahn–Teller insulating phase upon crossing an insulator-to-metal barrier with decreasing intermolecular distance (Fig. 1).[1] The normal state of these superconductors shows unconventional metallic behaviour as evidenced by magnetisation, nuclear magnetic resonance (NMR) and infrared (IR) spectroscopy measurements. The IR spectroscopic manifestation of this phase is the co-existence of the excitations of an extended electron system with the vibrations of a Jahn–Teller distorted molecular ion, the source of the distortion being localised electrons. Thus this phase can be termed a Jahn–Teller metal. Upon further decrease of the interfulleride distance, the localised nature of the electron system gradually disappears and a conventional Fermi liquid is created (Fig. 1). The various IR spectroscopic signatures of the fcc $Rb_xCs_{3-x}C_{60}$ series guide us through the evolution of the electronic structure upon changing the temperature and the chemical pressure. The localised nature can be followed by the presence of the Jahn–Teller-split vibrational lines, the metallic nature by the electronic background and the developing conventional electronic continuum by the appearance of asymmetric Fano lineshapes.

[1] R. H. Zadik, Y. Takabayashi, G. Klupp, R. H. Colman, A. Y. Ganin, A. Potočnik, P. Jeglič, D. Arčon, P. Matus, K. Kamarás, Y. Kasahara, Y. Iwasa, A. N. Fitch, Y. Ohishi, G. Garbarino, K. Kato, M. J. Rosseinsky, K. Prassides, "Optimized unconventional superconductivity in a molecular Jahn-Teller metal". Science Advances 1, e1500059/1-9 (2015).

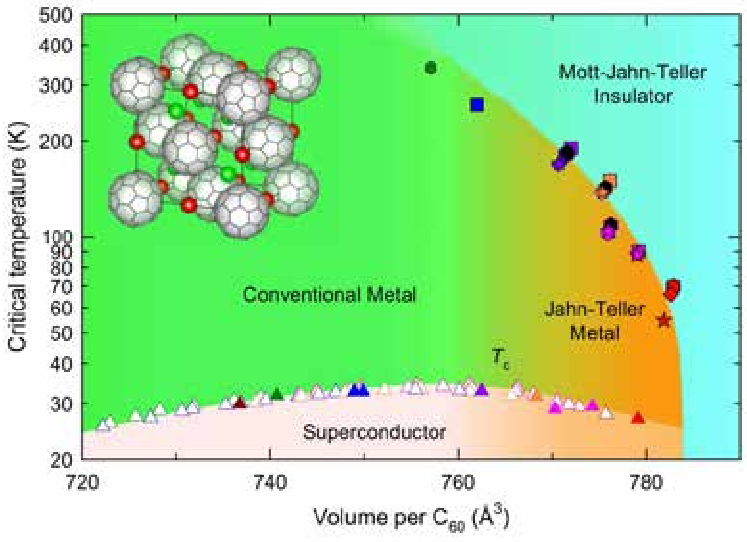


Figure 1: Electronic phase diagram of superconducting A_3C_{60} fullerides with their structure shown in the inset. Transition temperatures measured by X-ray diffraction, magnetization, NMR and IR set out regions of different electronic phases.[1]

PI-4

Charge dynamics in non-equilibrium state of dimer-Mott insulator β'-(BEDT-TTF)$_2$ICl$_2$ investigated by noise spectroscopy

M. Kurosu[1], B. Hartmann[2], J. Müller[2], S. Iguchi[1], H. Taniguchi[3], T. Sasaki[1]
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[2]*Goethe-University, Institute of Physics, Frankfurt, Germany;*
[3]*Saitama University, Department of Physics, Saitama-shi, Japan*

β'- (BEDT-TTF)$_2$ICl$_2$ is a quasi-two-dimensional molecular dimer Mott insulator with one hole carrier in the BEDT-TTF molecule dimer. Recently, Iguchi et al. reported that the temperature dependence of the dielectric constant shows a broad peak structure around 60 -80 K with frequency dispersion, and found an anisotropic glassy ferroelectricity by pyrocurrent measurements, which imply charge disproportionation resulting in an electric dipole in each dimer [1]. Although there is no signature on the breaking of inversion symmetry, nor the charge ordering in this system, local charge disproportionation in a dimer has been suggested by Raman scattering measurements in electric fields. Therefore, this polarized state is not due to spontaneous intra-dimer charge disproportionation, but is a metastable disproportionation induced by electric fields. Such polarized state has also been expected from theoretical studies on the charge degrees of freedom in a dimer Mott insulating phase [2, 3]. In this study, we conducted transport noise measurements to investigate the charge fluctuation dynamics related to the electric-field-induced metastable state. The transport noise was measured as the current fluctuations as a function of temperature. The normalized noise spectral density at 1 Hz (S_I/I^2) decreases with decreasing temperature from room temperature. We observed large exponential enhancement of S_I/I^2 below 200 K for sample 1 and 150 K for sample 2. In the same low temperature region, a non-linear conduction appears simultaneously with the noise enhancement. The temperature dependence of S_I/I^2 in the low temperature region obeys a theoretical expression for the transport noise in variable range hopping conductions. These results suggest that the charge hopping situation arises from the metastable charge disproportionation.

[1] S. Iguchi et al., Phys. Rev. B 87, 075107 (2013).
[2] M. Naka et al., J. Phys. Soc. Jpn. 79, 063707 (2010).
[3] C. Hotta, Phys. Rev. B 82, 241104(R) (2010).

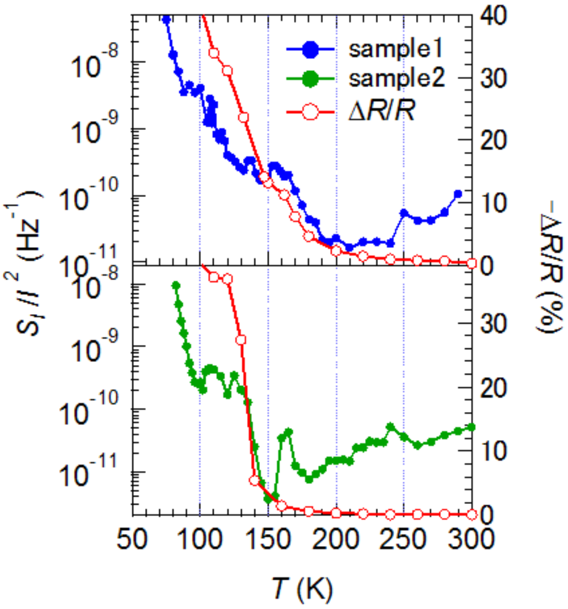

Figure 1: Temperature dependence of the normalized noise spectral density at 1 Hz and the rate of change of resistivity

Charge order in dimerized κ-(BEDT-TTF)$_2$Hg(SCN)$_2$X: Examining the magnetic anisotropy

T. Ivek[1], M. Herak[1], R. N. Lyubovskaya[2], N. Drichko[3], M. Dressel[3]
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[2]*Russian Academy of Sciences, Institute of Problems of Chemical Physics, Chernogolovka, Russia;*
[3]*Universität Stuttgart, 1. Physikalisches Institut, Germany*

Electronic multiferroicity is at the forefront of investigations in solid state physics due to the promise of practical applications as well as its fundamental character. Among many candidate materials there has been a recent increase of interest in the κ-(BEDT-TTF)$_2$CuX family of quasi-2D dimerized organic conductors thanks to the pronounced dielectric response found in the magnetically-ordered phase [1]. Interestingly, in these compounds optical and NMR spectroscopies do not find any charge disproportionation, which is a basic prerequisite for electric dipoles [2,3]. On the other hand, a mercury-based system κ-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl shows clear evidence of charge ordering at T_{CO} = 30 K [4]. Below that temperature, the ESR parameters change sharply and an antiferromagnetic-resonance-like response is found [5]. To reveal the nature of this novel low-temperature phase, we present a detailed investigation of magnetic anisotropy in κ-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl. The obtained data complements infrared and preliminary dielectric results. We contrast the results with sister compound κ-(BEDT-TTF)$_2$Hg(SCN)$_2$Br where charge order is absent, and discuss our findings in light of recent theoretical descriptions.

[1] P. Lunkenheimer et al., Nat. Mater. 11, 755–758 (2012).; S. Tomić et al., J. Phys.: Condens. Matter 25, 436004 (2013); M. Pinterić et al., Phys. Rev. B 90, 195139 (2014).
[2] K. Sedlmeier et al., Phys. Rev. B 86, 245103 (2012)
[3] Y. Shimizu et al., Phys. Rev. B 73, 140407 (2006).
[4] N. Drichko et al., Phys. Rev. B 89, 075133 (2014).
[5] S. Yasin et al., Physica B 407, 1689 (2012).

PI-6

Excitonic fluctuation in tilted Dirac electron system: A possible mechanism of low-temperature anomaly in α-ET salts

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In organic conductor α-(BEDT-TTF)$_2$I$_3$, a two-dimensional massless Dirac electron system is realized under hydrostatic pressure [1]. In contrast to graphene, its layered structure enables bulk-probing experiments such as nuclear magnetic resonance (NMR), inter-layer transport and specific heat measurements. Recently, a low-temperature anomaly was observed by NMR and in-plane resistance [2,3]; the latter shows an increase below 5~10K as the temperature is decreased while it is constant at higher temperatures. The 1/T$_1$T also turns to increase at low temperatures (below 5~10K). These behaviors cannot be understood by a non-interacting Dirac model, and it is speculated that Coulomb interaction plays some roles. Indeed, the long-range nature of the Coulomb interaction can be important in this system because of weak screening due to the vanishing density of states at the Dirac (band-degeneracy) points [4]. In this work, we study the effects of the long-range Coulomb interaction in a two-dimensional Dirac Hamiltonian with tilting (see Figure). By focusing on the fluctuations of the excitonic electron-hole condensation [5], we investigate a possible mechanism of the low-temperature increase of 1/T$_1$T under an in-plane magnetic field [2]. It is found that, under magnetic field and with the tilting of the Dirac cones, the leading instability of the excitonic condensation is in the inter-valley spin-triplet channel, and the fluctuations towards this state contribute to the increase of 1/T$_1$T.

[1] K. Kajita et al., J. Phys. Soc. Jpn. 83, 072002 (2014).
[2] Hirata Doctor thesis in The Univ. of Tokyo (2012).
[3] N. Tajima, Phys. Rev. Lett. 102, 176403 (2009).
[4] V. N. Kotov et al., Rev. Mod. Phys. 84, 1067 (2012).
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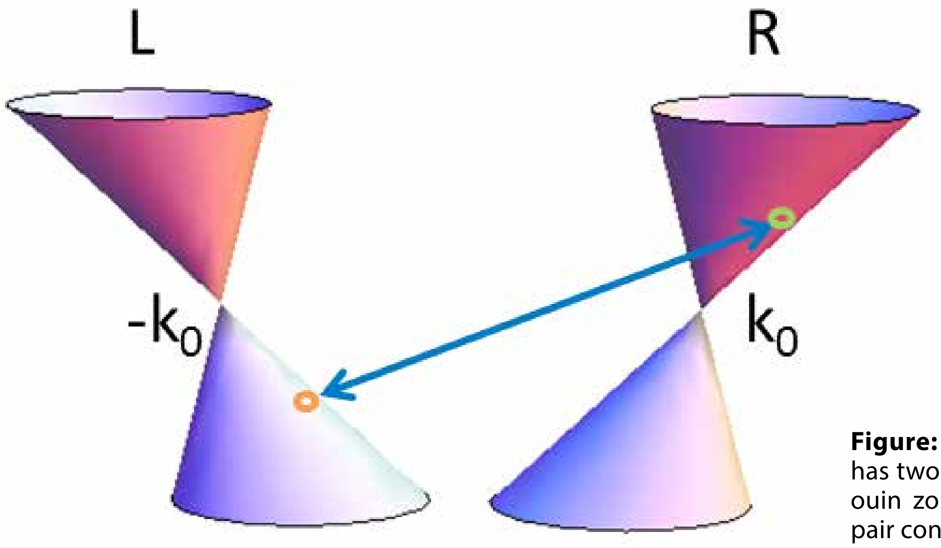

Figure: The band structure of the α-ET salt has two tilted Dirac cones in the first Brillouin zone. The inter-valley electron-hole pair considered here is shown.

Crystallization and vitrification in charge degrees of freedom in geometrically frustrated organic conductors

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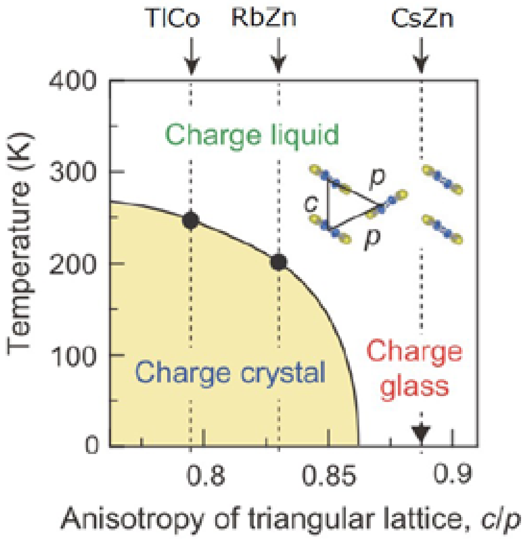
Upon cooling, liquids are normally transformed into solids which are long-range ordered below the melting point. However, the crystallization can be kinetically avoided by sufficiently rapid cooling and eventually glassy states can result generally in atomic / molecular configurational degrees of freedom. The glassy state is one of non-equilibrium states. Recently, several phenomena analogous to the structural glasses are found in the charge degrees of freedom in an organic conductor θ-(BEDT-TTF)$_2$X(SCN)$_4$ (X=CsZn, RbZn, TlCo)[1-3]. They exhibit "charge crystal" with a horizontal stripe-order at low-temperatures in equilibrium; when they are rapidly cooled, however, the first-order charge ordering transition is gone and a "charge glass state" shows up instead. The charge glass is characterized by slowing down of charge fluctuations on cooling, short-ranged charge-ordering domains and non-equilibrium aging behavior. The θ-type materials have anisotropic triangular lattices, which bear geometrical frustration against charge ordering in the 1/4-filled-band. In the present work, we have investigated the material dependence of the glass behavior and found a variation in the critical cooling-speed for vitrification, which indicates that stronger charge frustration leads to superior charge-glass former. In the symposium, we present comparatively the glass behavior of the three compounds.

[1] F. Kagawa, T. Sato et al., Nat. Phys. 9, 419 (2013).
[2] T. Sato, F. Kagawa et al., Phys. Rev. B 89, 121102(R) (2014).
[3] T. Sato, F. Kagawa et al., J. Phys. Soc. Jpn. 83, 083602 (2014).



θ-type phase diagram: electronic state is plotted with respect to the ratio of the two different triangular lattice parameters, c/p

PI-8

Pt-NMR study on $X[Pt(dmit)_2]_2$ system

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The characteristic band structure due to the narrow HOMO-LUMO gap and strong dimerization nature is realized in $X[Pd(dmit)_2]_2$ system. Most $Pd(dmit)_2$ compounds exhibit a (Mott) insulating state in the whole temperature range expected in a correlated half-filled system, because the intra-dimer transfer is considered to be much larger than inter-dimer one. Additionally, since the $[Pd(dmit)_2]_2$ dimers form a (quasi) triangular arrangements in the two dimensional layer, electronic correlation effect should competes and/or coexists with the frustration effect. Various unconventional physical behaviours observed in the members of this family should be related to the characteristic band structure and the frustration. On the other hand, the Pt-substituted system, $X[Pt(dmit)_2]_2$ is found to have relatively longer Pt-Pt distance compared to the Pd-Pd one in $Pd(dmit)_2$. Therefore the Pt system is expected to have smaller dimerization nature than the Pd system with similar crystal symmetry. In this system, metallic state can be expected because the antibonding HOMO and bonding LUMO bands may approach with each other with reducing dimerization gap. In fact, some Pt compounds behave metallic at high temperatures and they undergo a metal-insulator transition at low temperature. We performed 195Pt NMR measurements on $(Me_4P)[Pt(dmit)_2]_2$ to detect the electronic properties at the central Pt site. Almost temperature independent NMR shift and exponential decrease of the relaxation rate were observed below the metal insulator transition temperature. This reflects that the system has a non-magnetic ground state. The estimated spin gap is ~0.3 eV which is consistent with the transport measurements. Further measurements are under way

A systematic study of quasi-one-dimensional organic compounds α-(BPDT-TTF)$_2$X (X = ICl$_2$, AuCl$_2$) under pressure

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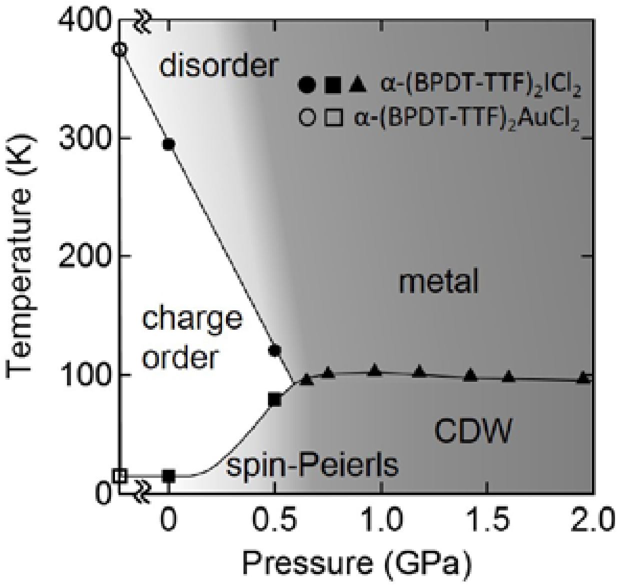
In one-dimensional (1D) systems, nesting of the 1D Fermi surface induces Peierls instability, which leads to a charge-density-wave (CDW) order. Contrastingly, in strongly correlated organic materials, Wigner-type charge ordering (CO) often emerges, where the off-site Coulomb interaction plays an important role. Here we show a comprehensive study of the electronic states under pressure in α-(BPDT-TTF)$_2$X (X = ICl$_2$,AuCl$_2$) with a quasi-1D 1/4-filled band. We find that the electronic states in α-(BPDT-TTF)$_2$X change from a Wigner-type CO state into a CDW state due to the increase of bandwidth when the physical pressure is applied. α-(BPDT-TTF)$_2$X forms a CO state already at room temperature owing to the strong off-site Coulomb repulsion. Whereas the temperature dependence of the resistivity at ambient pressure shows a clear phase transition at 300 K and 370 K for the X = ICl$_2$ and AuCl$_2$ salts, respectively, vibrational features observed in the infrared optical study have revealed that charge disproportionation remains in the higher temperature phase. These results indicate that this transition is a charge order-disorder transition. In the CO state, a wide band due to the electronic excitation is observed in the optical conductivity spectra. This band drastically shifts to lower frequencies with decreasing temperature. This behavior cannot be accounted for by a simple single excitation originating from the off-site Coulomb interaction, but rather by a domain wall excitation characteristic of a 1D CO system [1-3]. For both salts, at a lower temperature (~15 K), a spin-Peierls transition is observed. While the resistivity under pressure shows a suppression of the charge order-disorder transition, the spin-Peierls transition temperature is increased with increasing pressure. These two phase transitions coincide with each other at around 100 K and 0.6 GPa. The metal-insulator transition in the higher pressure region insensitive with pressure occurs due to a CDW instability.

[1]M. Mayr and P. Horsch, PRB 73, 195103 (2006)
[2]S. Fratini and J. Merino, PRB 75, 195103 (2007)
[3]A. Antal et al., PRB 87, 075118 (2013)



P-T phase diagram: P-T phase diagram of (BPDT-TTF)$_2$X. The open and closed symbols correspond to X=ICl$_2$ and AuCl$_2$, respectively. Circles, squares, and triangles indicate the charge order-disorder transition, the spin-Peierls transition, and the CDW transition, respectively.

PI-10

Observation of the charge-order gap in δ-$(ET)_4$[2,6-anthracene-bis(sulfonate)]·$(H_2O)_4$

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Recently, a new δ-ET salt with the 2,6-anthracene-bis(sulfonate) dianion and H_2O molecules was synthesized [1]. It is metallic at room temperature and exhibits a metal-insulator transition at T_{MI}=85 K. Below TMI we observe a charge disproportionation related with the progressive switching of bifurcated OH···O hydrogen bonds between the sulfonate moieties of anion and the water molecules. This salt shows how structural modifications within the anion layers directly influence the electronic properties of organic layers through hydrogen bonds. To gain more insight into the nature of phase transition, we have performed optical investigations for single crystals of the δ-$(ET)_4$[2,6-anthracene-bis(sulfonate)]·$(H_2O)_4$ salt in a wide spectral region from 25 to 18 000 cm^{-1}. The opening of a charge-order gap can be detected in the resistivity and FIR measurements. As the energy gap opens in the CO state, the FIR optical conductivity drops dramatically, and we were able to estimate the energy gap at $2\Delta \approx 200$ cm^{-1}. The optical conductivity reveals a low-energy collective mode around 70 cm^{-1}. At low temperature, a band at 600 cm^{-1} and a wide band at 2500 cm^{-1} are seen. We can associate the band in the MIR range with site-to-site transitions within a fluctuating CO pattern (transition between „Hubbard-like“ bands). The band at 600 cm^{-1} („charge fluctuation band“) is due to transitions between the density of states at the Fermi energy and the Hubbard-like bands. The presence of the latter feature proves that we observe CO fluctuations in our system.

[1] F. Camerel et al. Cryst. Growth. Des. 13 (2013) 5135. *This work was supported by the NSC (DEC-2012/04/M/ST3/00774) and by the Deutscher Akademischer Austausch Dienst (DAAD)*

PI-11

^{13}C NMR in organic conductor κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ by partial BEDT-STF substitution

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Quasi-two-dimensional organic conductor κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ shows no magnetic ordering at low temperature. The result is considered to be geometrical frustration of spins because triangular lattice consists of transfer integrals is nearly isotropic [1], whereas the competition between Mott localization and Anderson-like localization picture was proposed [2]. If the absence of magnetic ordering in the κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ is due to the frustration, the magnetic frustration should change by suppressing the geometrical frustration. One of the ways to distort the triangular lattice is the substitution with BEDT-STF. Transfer integrals around BEDT-STF are modified, which distorts the triangular lattice locally. As a result, the geometrical frustration is suppressed. Moreover the substitution introduces disorder into conduction layer. We performed ^{13}C NMR, electric conductivity, and static susceptibility measurements in order to elucidate electronic property in the κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, with the BEDT-STF substitution. We found that the static and dynamic susceptibility did not change even at the lowest temperature which is lower than one hundredth of the exchange interaction $J \simeq 250$ K as opposed to the great impurity substitution effect of the conductivity. We will report the results in more detail in this presentation.

[1] Y. Shimizu et al., Phys. Rev. Lett. 91, 107001 (2003).
[2] M. C. O. Aguiar et al., Phys. Rev. Lett. 102, 156402 (2009).

PI-12

Dilatometry studies focusing on the glass-like transition in the asymmetric quasi-1D organic salt δ-(EDT-TTF-CONMe$_2$)$_2$Br

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The organic charge-transfer salt δ-(EDT-TTF-CONMe$_2$)$_2$Br is a quasi-1D quarter-filled charge-ordered Mott insulator. Here we present measurements of the uniaxial thermal expansion coefficients αi along the i = a- and b-axis for temperatures 4.2 K ≤ T ≤ 200 K. Apart from the anisotropic lattice effects, a pronounced second-order structural phase transition from orthorhombic to monoclinic structure at ~ 190 K has been detected. A new feature has been observed in the temperature range 100-130 K, cf. Fig. 1, attributed to a glass-like transition which depends on the system's kinetics, i.e., warming and cooling rates across the transition. Such transition for an asymmetric quasi-1D system, including a point-like anion (Br$^-$), has not been revealed until now. The activation energy, derived from an Arrhenius plot, is found to be (3,602 ± 67) K, similar to the value obtained for κ-(ET)$_2$Cu[N(CN)$_2$]Br [1]. Such a glass-like transition in this material could be due to the vibrational motions of ethylene groups of the asymmetric EDT-TTF-CONMe$_2$ molecule. Our data lack any signatures of an antiferromagnetic transition, revealed at 12.5 K by NMR measurements [2], which might be due to the influence of the glass-like transition at higher temperature.

[1] P. Auban-Senzier et al., PRL 102, 257001 (09).
[2] J. Müller et al., PRB 65, 144521 (02).

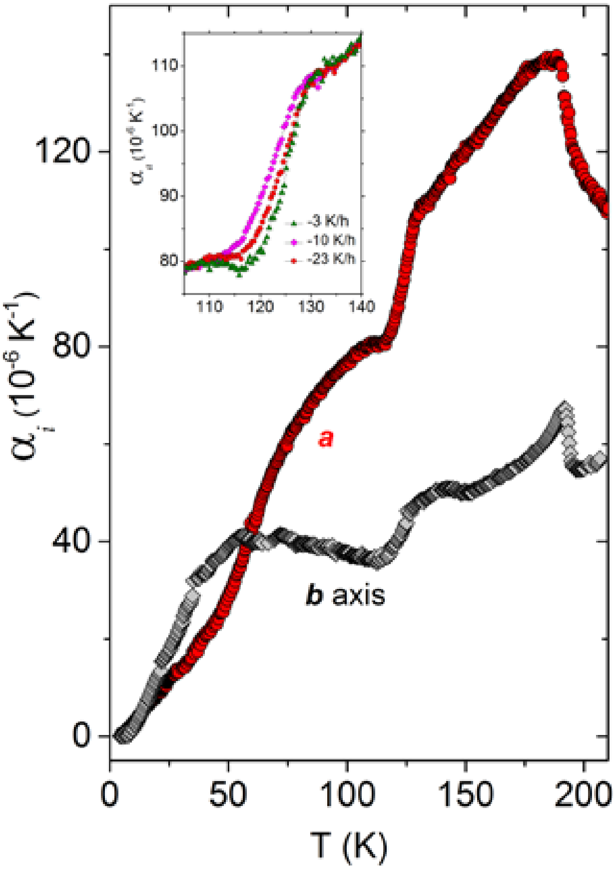

Figure: Temperature dependence of the uniaxial expansivities measured along the i = a and b-axis. Inset shows the expansivity along the a-axis across the glass-like transition for different cooling rates on expanded scales.

Optical investigation of $(TMTTF)_2X$ under pressure

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$(TMTTF)_2X$[1] are organic conductors which show low-dimensional electronic properties due to strong anisotropy arising from the crystal structure. Because of the much stronger overlap of the molecular orbitals in the stacking direction these materials are assumed to be quasi one-dimensional. This leads to a very rich phase diagram with charge order, spin peierls, antiferromagnetic, charge density wave and superconducting phases (see figure). This phase diagram was established by different techniques, such as NMR [2] and DC transport [3], by applying external hydrostatic pressure and investigating compounds with various anions. For the first time, we present optical investigations on the compounds $(TMTTF)_2AsF_6$ and $(TMTTF)_2SbF_6$ under pressure. In this work we used a piston pressure cell [4] with diamond as optical window and daphne oil as the pressure medium (see figure). The accessible range of pressure is 11 kbar and the lowest reachable temperature is 7 K, which covers the highlighted range in the phase diagram. The biggest advantage of optical measurements is the possibility to directly access the amount of charge per molecule via polarization-dependent vibrational spectroscopy on charge-sensitive modes in the mid-infrared.

[1] M. Dressel et al, Crystals 2012, 2, 528-578
[2] F. Zamborszky et al., Phys. Rev .B 66, (2002) 081103
[3] P. Auban-Senzier et al., J. Phys. IV France114 (2004) 41
[4] R. Beyer, M.Dressel, to be published

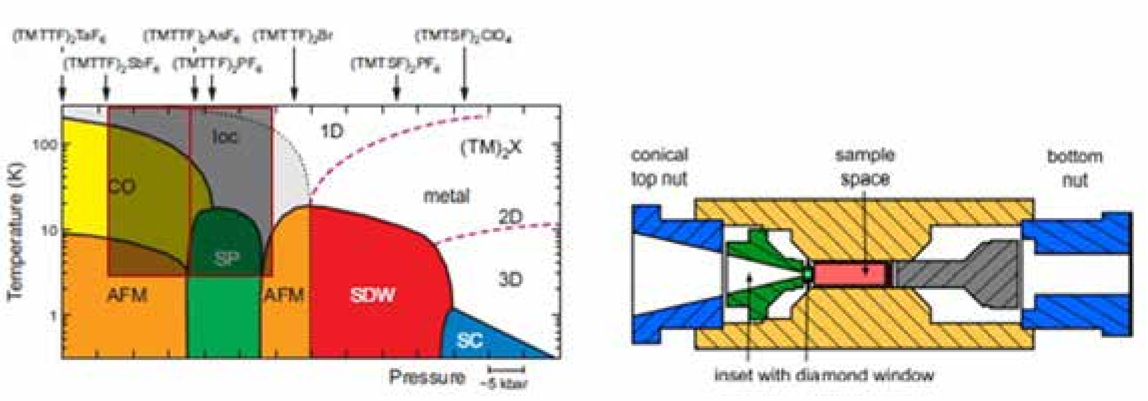

Figure: The phase diagram of the quasi one-dimensional TMTTF and TMTSF salts with accessible range for measurements. Schematic view of the piston pressure cell.

PI-14

Anisotropic reshaping of a tilted Dirac cone in the organic salt α (BEDT TTF)$_2$I$_3$ probed by NMR

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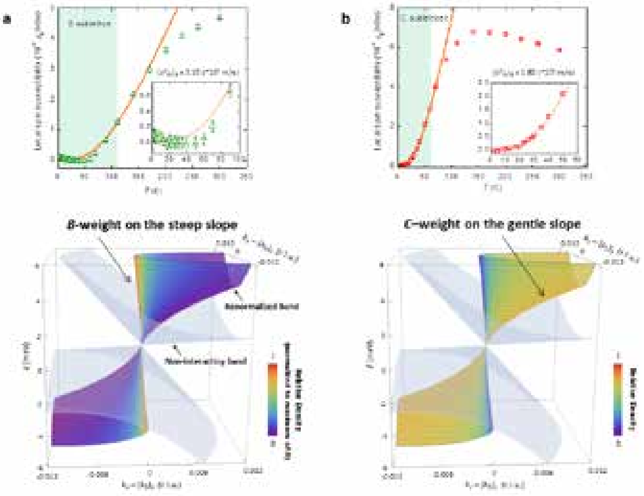
The organic salt α-(BEDT TTF)$_2$I$_3$ is a candidate for a Dirac cone system under pressure that is characterized by the four sublattices in the unit cell (molecules A, A', B and C) and the strong tilt of the cone [1,2]. The band crossing points (the Dirac points) locate in the upper two bands in the vicinity of the Fermi level, around which the electronic states are governed by an effective Weyl Hamiltonian and mimic relativistic massless fermions as in graphene [3]. Reflecting the 4-site nature, the states around the Dirac point are described by a superposition of the four orbital states that reflects the relative weights on the four different sublattices. Theory suggests that the density on a particular sublattice varies as a function of the k vector around the Dirac point such that the density for the B and C sites is maximized in the different regions on the cone, i.e., in the steep slope and the gentle slope, respectively [2]. Using NMR that is able to probe the different sublattices, we experimentally demonstrate this effective "sublattice to k" correspondence. The Knight shift measurement reveals deviations from the T-linear behavior expected for the linear dispersion [4] that exhibit distinct temperature scales for the different sublattices (Figure). Assuming a logarithmic enhancement of the Fermi velocity around the Dirac point and a Hubbard-U like suppression at the higher energy due to the long-range and short-range parts of the Coulomb interaction, respectively, we show that the data can be properly fitted using different velocities for the different sublattices, demonstrating an anisotropic reshaping of the cone (Figure) [5]. In the talk, I will discuss the long-wave-length nature of this renormalization by making comparisons of the shift and relaxation rate data.

[1] N. Tajima et al., Phys. Rev. Lett. 102, 176403 (2009).
[2] S. Katayama et al., Eur. Phys. J. B 67, 139 (2009).
[3] M. Goerbig et al., Phys. Rev. B 78, 045415 (2008).
[4] B. Dóra et al., Phys. Rev. Lett. 102, 197602 (2009).
[5] H. Isobe et al. J. Phys. Soc. Jpn. 81, 113704 (2012).

We thank Dr. N. Suzumura, A. Kobayashi, N. Nagaosa, H. Fukuyama, N. Tajima, Y. Tokunaga and M-H. Julien for fruitful discussions. Above all, special thanks go to Dr. M. O. Goerbig and M. Horvatić for helping us to interpret the data and to make the model fitting analyses.



Reshaping of tilted cone: The relative weight difference in the k-space for the different sublattices (B and C) enables one to resolve the interaction-induced renormalization effects on the different slopes separately. (Solid curves in the top panel stand for the fit).

Negative-temperature state and phase separation induced by symmetric monocycle optical pulse in extended Hubbard models

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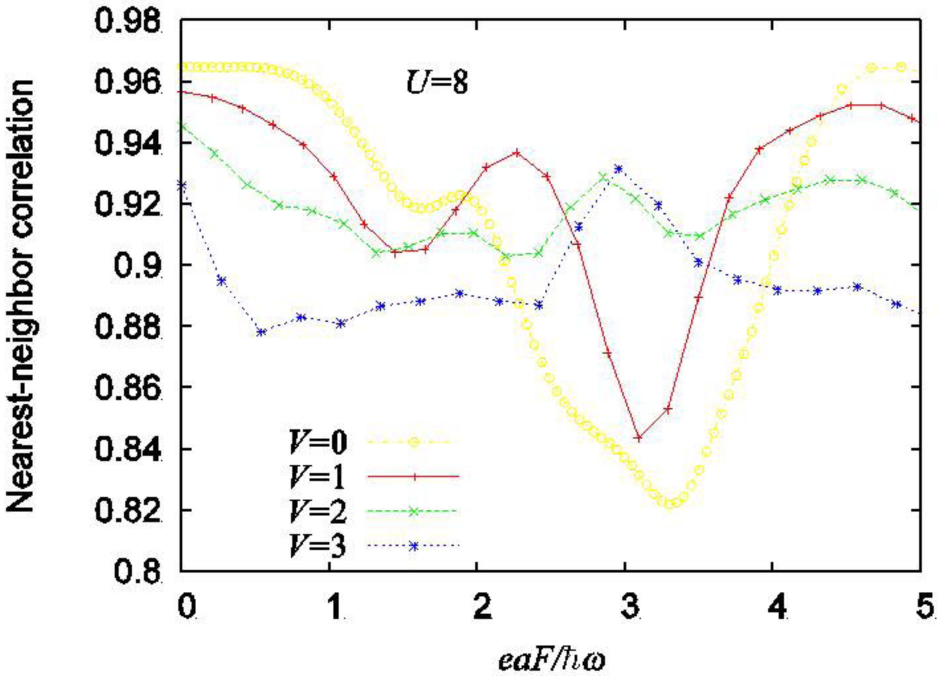
Many-electron dynamics induced by an optical pulse of large electric field amplitude are theoretically investigated in one- and two-dimensional correlated electron and electron-phonon systems using the exact diagonalization method. A ``negative-temperature“ state and inversion of electron-electron and electron-phonon interactions (e.g., change from repulsive interactions to effective attractive interactions) are induced even by a symmetric monocycle pulse, where the time integral of the electric field is zero and the dynamical phase shift vanishes. They are realized for a wide region of the field amplitude and the excitation energy, in the band-insulator [1] did not realize a negative-temperature state by a symmetric monocycle pulse in the dynamical mean-field theory. In the Mott-insulator and charge-ordered-insulator phases on regular lattices (i.e., without dimerization) with a nonnegligible intersite repulsive interaction, the corresponding effective attraction causes phase separation between charge-rich and charge-poor sites. Photoinduced phase separation is much more feasible than photoinduced superconductivity proposed by Tsuji et al. in the Hubbard model [2].

[1] J. Phys. Soc. Jpn. 84, 054702 (2015)], Mott-insulator and charge-ordered-insulator phases. This work suggests that why the previous studies by Tsuji et al. [Phys. Rev. B 85, 155124 (2012)
[2] Phys. Rev. Lett. 106, 236401 (2011)



Photoinduced correlation: Time-averaged nearest-neighbor charge-density correlation as a function of field amplitude/excitation energy for different values of nearest-neighbor repulsion V. The on-site repulsion is U=8. The excitation energy is resonant with the optical gap

PI-16

Photo-induced phase separation in metallic phase of the organic compound κ-$(ET)_2Cu(NCS)_2$

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Photo-induced phase separation (PIPS) has been widely studied in various systems, such as a polymer blend, two-component nanoparticles and spin-crossover solid. For conducting electrons in solids, it has been reported that metallic-insulating PIPS occurs in the organic compound κ-$(ET)_2Cu[N(CN)_2]$ Br (κ-Br) [1]. However, although the PIPS was observed below the glasslike structural transition temperature ~ 80 K, it has not been confirmed whether the structural transition is essential in the PIPS or not. In this study, to obtain convincing evidence of such the structural effect on the PIPS, we have carried out pump-probe spectroscopy with different probe polarization in the κ-$(ET)_2Cu(NCS)_2$, which has similar crystal structure to κ-Br and shows glasslike structural transition at 70 K [2]. The optical measurements were performed using 120 fs pulses centered at 400 nm for a pump (134 μJ/cm^2) and 800 nm for a probe from a cavity-dumped Ti:sapphire oscillator with a repetition rate 50 kHz. The pump and probe beams were coaxially overlapped and irradiated perpendicular to the b-c plane. Figure 1(a) shows the polar plots of maximum values of the transient reflectivity ΔR/R for the probe-polarization angle. For 72 K, the signal is almost isotropic while it is anisotropic for 66 K. Moreover, the anisotropic (a_{max}) component, which is decomposed by fitting the data with a cosine function, shows steep increase at 70 K with decreasing temperature as shown in Fig. 1(b), indicating formation of an insulating gap. Since anisotropic ΔR/R for the probe arises from structural and/or electronic phase transition [3,4], it is consistent with the glasslike transition. The data will be discussed in detail at the conference.

[1] Y. Toda et al., Phys. Rev. Lett. 107, 227002 (2011)
[2] J. Muller et al., Phys. Rev. B 65, 144521 (2002)
[3] Y. Toda et al., Phys. Rev. B 90, 094513 (2014)
[4] S. Tsuchiya et al., submitted

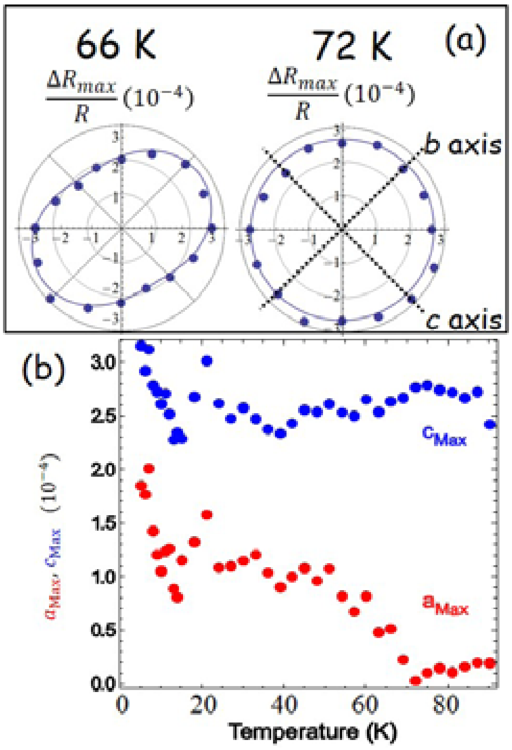

Figure 1: (a) Polar plots of the maximum values of transients for 66 K and 72 K. (b) Temperature dependence of magnitude of the isotropic (cmax) and anisotropic (amax) components.

PI-17

Charge-vibration dynamics driven by 6 fs, 1.3-cycle infrared pulses in dimer-Mott insulators

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[2]*University of Yamanashi, Department of Applied Chemistry, Japan;*
[3]*Institute for Materials Research, Tohoku University, Sendai, Japan*

A series of κ-type BEDT-TTF salts has been known as a phase-rich compound exhibiting Mott transition, superconducting, spin-liquid and electronic ferroelectricity.... by modulating U_{dim}/t (U_{dim}: effective on-site Coulomb energy, t: interdimer transfer energy). Such flexible electronic structure, enables us to expect ultrafast and dramatic responses by light stimulation. We have reported efficient (>100 molecules/photon) photoinduced insulator-to-metal transition (PIMT) in κ-(d-BEDT-TTF)$_2$Cu[N(CN)$_2$]Br [1]. However, the excited state dynamics remains unclear in the time scale of $\hbar/t$ and intra- / inter- molecular vibration interacting with charge. In this study, an early stage of the PIMT has been investigated by 6 fs (1.3 optical cycles) infrared pulses. Figure shows the transient reflectivity ($\Delta R/R$) spectra measured by 100 fs and 6 fs pulses. Before the appearance of the Drude-like response (< 0.4 eV) at 1 ps ((b) *), we observed reflectivity increase (> 0.65 eV) reflecting the excited state dynamics. The peak of $\Delta R/R$ moves toward high-energy (< 30 fs: (c) **) and splits into two peaks (30 - 100 fs: (c) ***) with the oscillating structure (1300 cm^{-1}). Even at 10 fs after the excitation, the electronic excited state has already been coupled with intramolecular vibration (ν3 mode). Then, intermolecular coherent motion (27, 60 cm^{-1}) drives the metallization in the time scale of ~1ps[1]. These results show that the PIMT is driven by interactions between charge, intramolecular vibration, and intermolecular (intradimer) oscillation. Hierarchy of dimer-Mott system – charges, molecules, dimers interacting with each other – plays an essential role in the efficient PIMT. That is contrast to the melting of charge order driven by electronic interaction in α-(BEDT-TTF)$_2$I$_3$ [2].

[1] Kawakami, Iwai et al., Phys. Rev. Lett. 103, 066403 (2009)
[2] Kawakami, Iwai et al., Phys. Rev. Lett. 105, 246402 (2010)

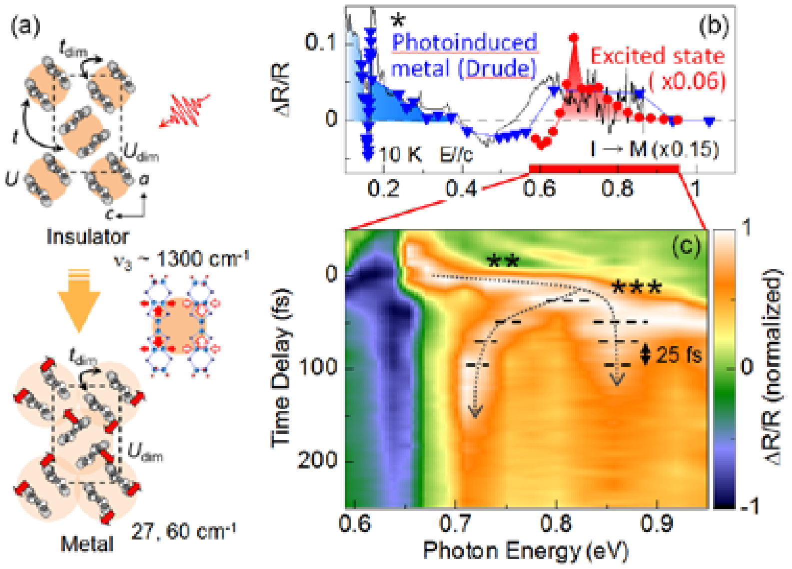

Figure: (a) Schematic illustration of PIMT. (b) Transient reflectivity spectra. (c) Temporal change of normalized spectrum

PI-18

Observation of terahertz excitations accompanied by charge fluctuations near the metal-insulator transition in α-(ET)$_2$I$_3$

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[2]*Okayama University of Science, Department of Applied Physics, Japan;*
[3]*Toyota Physical and Chemical Research Institute, Nagakute, Japan;*
[4]*Nagoya University, Department of Applied Physics, Japan*

Terahertz (THz) response near the metal-insulator transition in a-(ET)$_2$I$_3$ has been investigated by steady-state spectroscopy and subpicosecond time-resolved measurements upon photoexcitation. THz conductivity s of the compound below optical gap ~ 600 cm^{-1} is characterized by several phonon structures below the charge-ordering(CO) temperature T_{CO} = 135 K, and by broad increase or closing of the gap above T_{CO} reflecting the metal transition[1-3], as shown in Fig. (a). At 131 K ~ T_{CO} in contrast, we identified peak structures at ~ 50 and ~ 100 cm^{-1} which are absent well below and above T_{CO}. The observed modes characteristic to the vicinity of T_{CO} should be attributable to the „intermediate“ state between the metal and the insulator(CO) phases. Such in-gap excitations with relatively broad widths suggest contribution of strongly-correlated electrons, namely, collective excitations of charge fluctuations at the phase boundary [Fig. (c)]. In fact, these modes can also be activated by injecting metallic states into the CO phase via photoinduced phase transition. Figure (b) shows transient differential spectra of s after the photoexcitation at 0.89 eV(charge-transfer excitation). The observed broad increase at the delay time t = 0 ps(immediately after the excitation) and its disappearance at t = 10 ps corresponds to the formation and the relaxation of the photoinduced metal, respectively [2]. At t = 1 ps during the relaxation process, in contrast, where the metal and the CO states presumably coexist, peaks emerged at wavenumbers coincident with the aforementioned peaks observed near TCO, ~ 50 and ~ 100 cm^{-1}, indicating the common origin: the charge fluctuations at the phase boundary [Fig. (c)]. In the presentation, cooperative and critical behaviors of them will also be discussed.

[1] Y. Yue, K. Yamamoto, K. Yakushi et al., Phys. Rev. B 82, 075134 (2010).
[2] H. Nakaya, H. Itoh, S. Iwai et al., Phys. Rev. B 81, 155111 (2010).
[3] T. Ivek, M. Dressel et al., Phys. Rev. B 83, 165128 (2011).

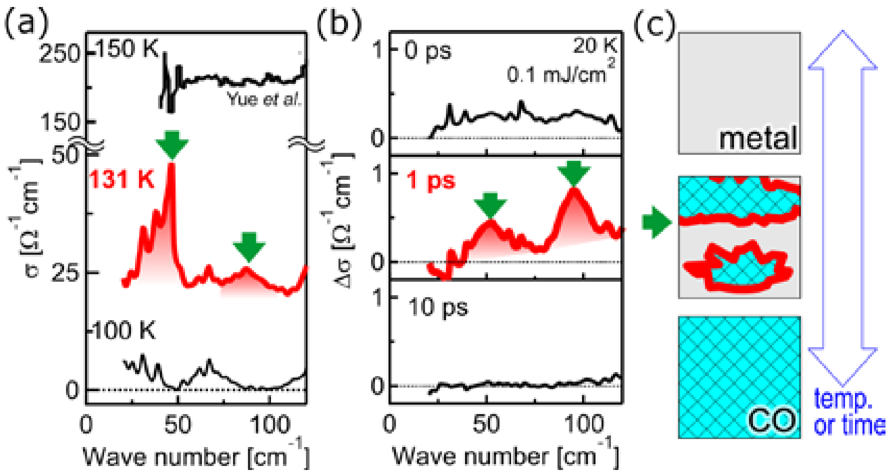

Figure: (a) Temperature dependence of steady-state conductivity spectra. (b) Transient differential spectra upon photoexcitation. (c) Schematics of metal-insulator phase boundaries.

PI-19

Ultrafast optical response of the metallic phase of α-$(ET)_2I_3$ induced by 7 fs infrared pulse

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Photoinduced insulator-to-metal transitions (PIMT) has been extensively studied in Mott insulators and charge ordered (CO) insulators. We have investigated the photoinduced CO melting in α-$(ET)_2I_3$ by using various type of ultrafast measurements [1-3]. On the other hand, there are few studies on the photoinduced effects in the metallic phase of organic conductors. Our fundamental question is that „Can we realize a reverse process of the PIMT, i.e, the photoinduced metal-to-CO insulator transition?". In this study, to answer the above question, we performed pump-probe spectroscopy using infrared 1.5 cycle (7 fs) pulse with strong electric field [4]. Fig. (a) shows the temporal change of the transient reflectivity ($\Delta R/R$) spectrum at 138 K ($> T_{CO} = 135$ K), plotted as a two-dimensional (delay time (td) vs. detection energy) map. Immediately after the irradiation of pulse with 9.3 MV/cm, reflectivity increase (> 0.6 eV) indicating of the metal-to-insulator change was observed (td < 50 fs). Spectral intensity is modulated with a period of 20 fs (red dotted line in Fig. (a) and Fig. (b)). Then, the $\Delta R/R$ spectrum changes to the shape reflecting the increase of electron/lattice temperatures (td > 50 fs). The intense 20 fs oscillation can be attributed to the CO gap oscillation showing that the CO gap opens in the metallic phase. The mechanism of such charge localization induced by strong light field will be discussed in terms of dynamical localization [5] that is the reduction of the intersite transfer integral t by the strong high-frequency field.

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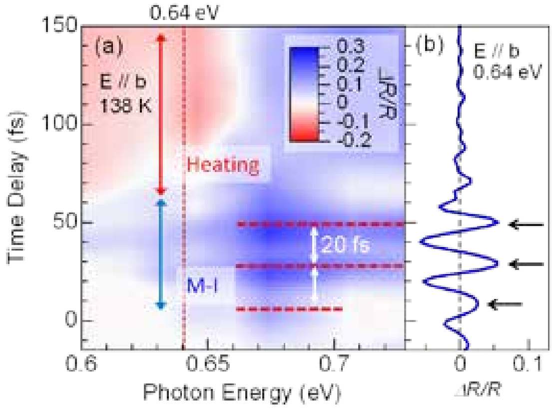

Figure: (a) Temporal change of the transient reflectivity spectrum after irradiation of 1.5 cycle pulse with 9.3 MV/cm. (b) Oscillating component of the time profile at 0.64 eV.

PI-20

Photocontrol of magnetism above 77 K in nanoscaled heterostructures of cyanometallate coordination networks: Mechanism and Limits

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Using nanometer-sized heterostructures of cyanometallate coordination networks, specifially core@shell nanoparticles of CoFe@CrCr-PBA (PBA = Prussian blue analogues), irradiation by white light at 80 K modifies the magnetic response, and these changes remain intact and persist without continued irradiation to nominally 125 K [1]. Preliminary pressure studies indicate the photoinduced changes can be maintained up to 200 K, the transition temperature of the ferromagnetic CrCr-PBA component. The effect, which we first reported up to 70 K [2,3], arises from thermally induced interface strain, which is relaxed by irradiation of the photactive consituent, CoFe-PBA. The ferromagnetic domains in the strained interface region are affected and generate the persistent changes of the magnetism. Our understanding of this photo-magnetostructural mechanism enabled us to extend the phenomenon to include photoactive spin-crossover systems [4] and other ferromagnetic PBAs [5]. The potential path to higher temperatures will be sketched.

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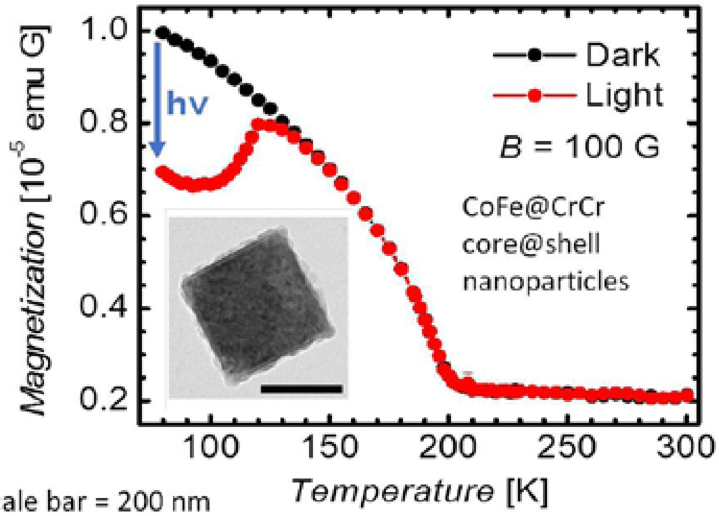

scale bar = 200 nm

Figure 1: Temperature dependence of the field-cooled magnetization without irradiation, „Dark“ data, and after irradiation at 80 K with the light switched-off for the „Light“ data.

PI-21

Shear Stress Effect on Photochromism of Diarylethene CMTE

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We have been interested in the shear stress effects on the photochromic compounds spiropyrans and diarylethenes, which exhibit the isomerization between the open- and closed-ring isomers by the irradiation of visible and UV light. Among these compounds, diarylethene is one of the most promoting materials [1]. Because they have exhibited the stable photochromic behavior in the single crystalline phase with thermally irreversible and fatigue-resistant properties. In this study, we have focused our attention on the diarylethene derivative, cis-1,2-dicyano-1,2-bis(2,4,5-trimethyl-3-thienyl)ethane, abbreviated as CMTE [2]. The *in situ* observations under a microscope with spectroscopic measurements have been carried out under shear stress using the DAC-type pressure cell with rotating mechanism. As the results, the combination of shear stress and visible light around 500 nm could induce the photoisomerization from the open-ring isomer to the closed-ring isomer. It is well-known for diarylethene CMTE crystal that the photochromism was caused by UV light of wavelength shorter than 390 nm. Therefore, the present work shows that shear stress could adjust the wavelength for the photochromism from ultraviolet light region to visible light around 500 nm. Our previous studies on nitrospiropyran have suggested that the shear stress could induce the chromic behavior from SP to merocyanine [3]. We present the results of CMTE as compared with spiropyrans [4].

The present work was supported by a Grant-in-Aid for Scientific Research (25410101) from the Ministry of Education, Culture, Sports, Science and Technology, Japan.

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PI-22

Photo-induced superconductivity in κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br utilizing a photo-active electric double layer

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Manipulating the electronic phases of a condensed matter by external stimuli is one of the hottest topics in the field of modern electronics. Strongly-correlated materials are good candidates for such phase transition devices. Recently, we have demonstrated a field-induced superconductivity in κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br (κ-Br). This is the first observation of field-induced superconductivity in an organic field-effect transistor [1]. In this study, we fabricated novel photo-active superconducting devices by laminating κ-Br on Al$_2$O$_3$/Nb-SrTiO$_3$ substrates coated with self-assembled monolayer of spiropyran-derivatives (SP-SAM) [2]. Because of the weak tensile strain-effect from the substrate, the initial resistances for the devices showed insulating properties. After UV irradiation, however, superconducting transition at 7.4 K was clearly observed, and remained even after the irradiation was stopped. The resistance recovered to nearly the initial value by visible light irradiation, showing a reversible switching capability. From our previous measurements, k-Br is known to evoke superconductivity by electrostatic carrier doping. The above reversible switching of superconductivity should also be due to a carrier doping by the formation of electric-double-layer (EDL) at the interface. Spiropyrans can switch between a non-ionic isomer and a zwitterionic isomer when triggered by light-irradiation with different wavelengths, resulting in a significant change in the electric dipole moment. Reversible changes in dipole moment of SP-SAM produced two distinct electric fields between the κ-Br and the substrates that created electrostatically doped carriers. Thus, superconductivity could be switched by photo-irradiation by photo-induced EDL formation.

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PI-23

Evidence for pre-formed Cooper pairs in the pseudogap phase of slightly underdoped $NdBa_2Cu_3O_{6+x}$

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In the last years ultrafast experiments have contributed to shed new light on high-temperature superconductivity. In particular, tailored excitation in the mid-infrared spectral range was demonstrated to suppress competing structural and electronic orders and to promote a highly coherent state in several underdoped cuprates [1-4]. In $YBa_2Cu_3O_{6+x}$ this transient state was found to persist up to room temperature, evidenced by the enhancement of the superfluid strength in the THz response. The question whether the high coherence is the signature of a perfect conducting or of an exotic superconducting state at nonequilibrium still remains open. Here, we address this problem from a spectroscopic point of view, by investigating a slightly underdoped sample of $NdBa_2Cu_3O_{6+x}$ through ultrafast spectroscopy in the optical regime. The use of a broadband detection scheme enables us to reveal evidence for quasiparticle (QP) excitation in a wide range of temperatures up to the pseudogap temperature scale T*. The existence of a QP spectral signature in the pseudogap phase, together with its peculiar temporal evolution and temperature dependence, can be directly related to the presence of a pairing gap for QP excitation. This observation leads to the hypothesis that the selective melting of a competing order using intense resonant mid-infrared pulses can establish coherence in pre-formed Cooper pairs underlying the pseudogap phase [5].

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PI-24

Random-walk annihilation process of photo-induced neutral-ionic domain walls in TTF-CA

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The charge and lattice dynamics at the neutral-ionic phase transition in the organic mixed-stack compound TTF-CA was measured by time-resolved Fourier-transform infrared spectroscopy in the microsecond time range. From the comparison of steady-state and time-resolved spectra we conclude that neutral, non-dimerized metastable domains are created after photoexcitation. The time evolution of the reflectivity reveals a stretched exponential decay, which can be fitted by the Kohlrausch-Williams-Watts-function. The change in reflectivity depends linearly on the laser intensity for all temperatures, but decreases with temperature in a nonlinear fashion. The observed time profile can be explained by a random-walk annihilation process of the generated neutral-ionic domain walls. From our comprehensive time-resolved infrared study and the random-walk model we can extend the known picture [1] and draw now a more detailed version of the domain size as a function of temperature and especially laser intensity. Close to the phase transition, large domains are formed (panels a and b) due to the valence instability. We also assert that the merger and interaction of the induced domains play an important role for the formation of the macroscopic domains and deduce from the model with decreasing laser intensity, the average domain size decreases. At lower temperatures the ionic phase is more robust; the average domain size is much smaller and changes less with laser intensity (panels c and d). We conclude that the random walk of the neutral-ionic domain walls is the dominant factor for the relaxation of the metastable domains in the temperature range considered [2].

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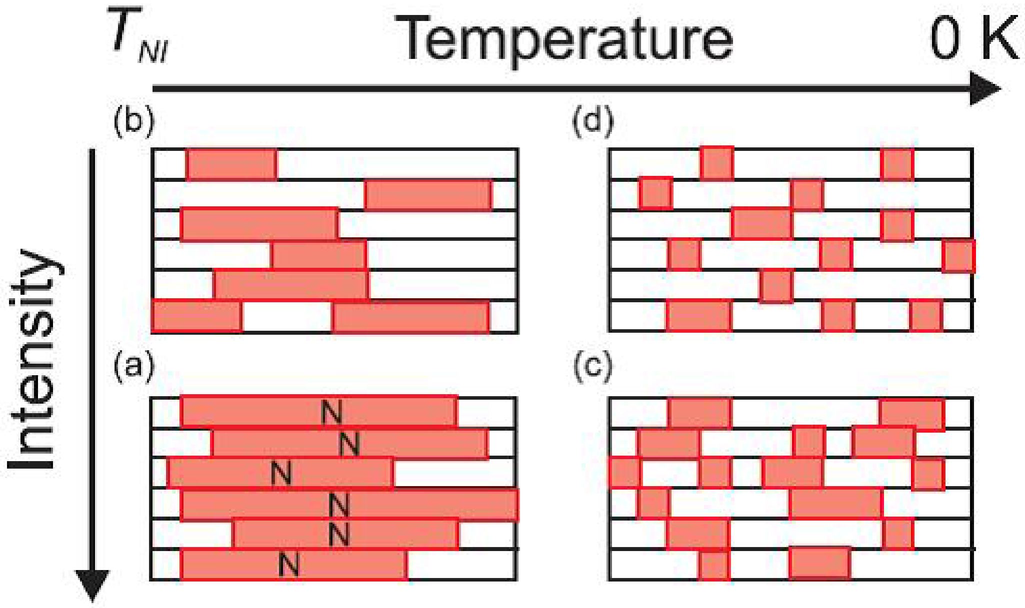

PI-25

Quantum and classical magnetoresistance oscillations in the organic superconductor κ-$(BETS)_2Mn[N(CN)_2]_3$ in the proximity of the Mott transition

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One of the most interesting of recently synthesized crystalline organic conductors is the bifunctional layered compound κ-$(BETS)_2Mn[N(CN)_{2]3}$ [1]. On the one hand, it exhibits a nontrivial interplay between the magnetism of paramagnetic Mn^{2+} ions in the insulating anion layers and itinerant spins of the conduction system in the radical cation BETS layers, involved in a metal-insulator transition at ~25 K [2,3]. On the other hand, it displays a competition and coexistence of the magnetically ordered insulating and superconducting states [1,4] typical of strongly correlated materials of current interest such as high-T_c superconductors. We have carried out high-field magnetoresistance studies of κ-$(BETS)_2Mn[N(CN)_{2]3}$ under pressure $P \approx 1.5$ kbar. This pressure places the compound into the metallic domain of the T- P phase diagram but rather close to the Mott-insulating region. We have found both Shubnikov-de Haas (SdH) oscillations and angle dependent magnetoresistance oscillations which provided a direct access to the Fermi surface of the compound. The SdH oscillations contain two frequencies: F_1= 1135 T and F_2=4215 T. The first one corresponds to the area of the closed pocket of the Fermi-surface, whereas the second one corresponds to the area of the Brillouin zone and is due to the magnetic breakdown effect. Both oscillation components are characterized by high cyclotron masses $5.7m_0$ and $7.0m_0$, respectively (m0 is the free electron mass), indicating a strong influence of electron correlations. The angular dependence of SdH oscillations is strongly suggestive of the spin-dependent out-of-plane transport in high magnetic fields, i.e. the tunneling probability through the anion layer with polarized Mn^{2+} anions seems to be considerably different for the carriers with the spins up and down.

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PI-26

Correlation between charge density wave and superconductivity in 2H-NbSe$_2$

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2H-NbSe$_2$ is a two-dimensional hexagonal layered compound exhibits Charge Density Wave (CDW) state at T_c = 35K followed by a superconducting (SC) state at T_s = 7.4K in high purity single crystal. The superconducting state of 2H-NbSe$_2$ belongs to type-II superconductor and we have found the anisotropic upper critical field ~ 3 around t = 0.81 where $t = T/T_s$ from parallel and perpendicular to the layer of NbSe$_2$ upper critical field. These an-isotropic upper critical fields are explained in terms of coherence length. However $H_{c2}^{t}(0)$ and $H_{c2}^{l}(0)$ were calculated ~ 2.68T and 8.15T respectively from the relation $H_{c2}(T)=H_{c2}(0)[1-t^2]$ at t = 0.84. This superconducting transition temperature was also found significantly reduced in 2H-Ga$_x$NbSe$_2$ and 2H-Nb$_{1-x}$V$_x$Se$_2$. Besides this, a new analytical formula for superconducting transition temperature $T_s = -(V_c/V_s)(\hbar w_s/\hbar w_c)T_c+(1-V_c/V_s)(1.13\hbar w_s/k_B)$ is derived from equations of BCS and Peierl's instability for 2H-NbSe$_2$ at constant normal state density of states (DOS) associated with T_c. It depends on phonon mode, effective interaction and CDW transition temperature. The pressure effect data of C. W. Chu et. al. is well fitted by the above analytical expression.

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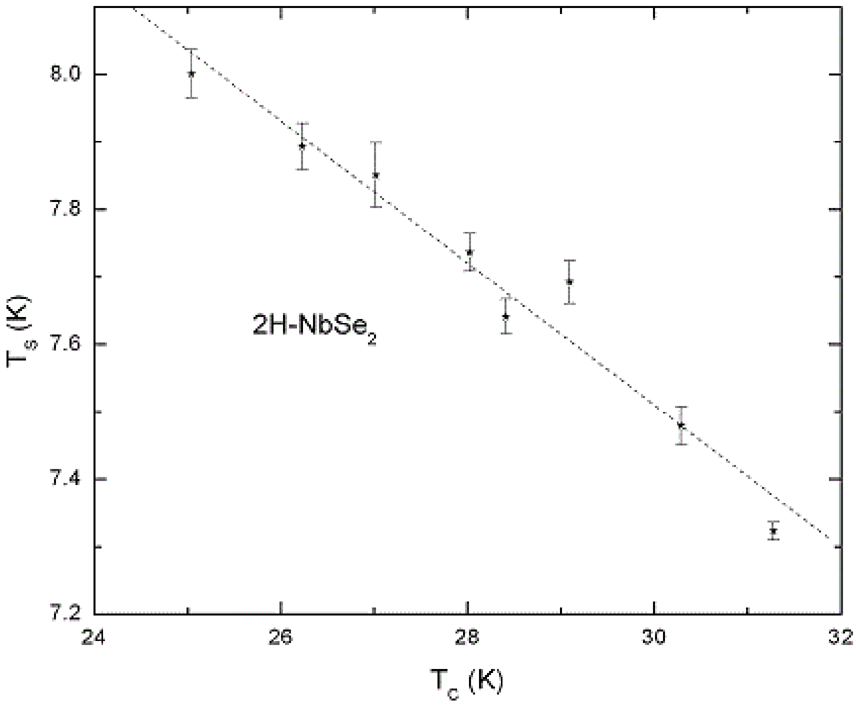

Figure 1: The pressure effect T_s and T_c data of 2H-NbSe$_2$ single crystal fitted by $T_s = -mT_c + C$

PI-27

Competing charge-ordering in two-dimesional organic metals β‘‘-(BEDT-TTF)$_2$X

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Organic superconductors are characterized by the interplay of different interactions (electron-electron, electron-phonon...), originating complex phase diagrams where weak additional forces may give rise to competing instabilities. In proximity of these instabilities fluctuations can occur, and in particular fluctuations of charge-order (CO) state have been invoked as the mediator of pairing mechanism in two-dimensional organic superconductors.

Here we present an exhaustive optical investigation of CO fluctuations in the series of isostructural salts b´´-(BEDT-TTF)$_2$X, among which the salt with X=SF$_5$CH2CF$_2$SO$_3$ is superconductor with T$_c$ = 5 K. CO instabilities and their dynamical properties have been studied through infrared and Raman spectroscopy. The vibrational modes most sensitive to molecular charge have been identified, and modeled though a modified “two states jump” Kubo model. In this way we have been able to characterize the CO amplitude and dynamics. In particular, in the superconductor salt, we have found an intriguing coexistence of static and dynamic CO states with nearly orthogonal arrangements, which could be relevant to the development of superconductivity. In fact the coexistence, which persists down to the lowest measured temperature of 4 K, is likely due to the competition of two different interactions driving towards different CO states. Combining these findings on the series of b´´-(BEDT-TTF)$_2$X salts with other experimental and theoretical studies, we propose a phase diagram for quarter-filled two-dimensional organic systems and discuss the nature and the origin of the different CO states.

PI-28

Non-trivial charge ordering near superconducting transition in β‘‘-$(BEDT\text{-}TTF)_4[(H_3O)Ga(C_2O_4)_3]\bullet C_6H_5NO_2$

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Unconventional superconductivity near charge ordered state demonstrates intriguing physical properties. In a organic superconductorβ‘‘-$(BEDT\text{-}TTF)_4[(H_3O)Ga(C_2O_4)_3]\bullet C_6H_5NO_2$, which shows a charge instability at 8.5 K and superconductivity at 7 K, the extremely high upper critical field was observed [1]. The robustness of superconductivity in high magnetic field might be related to the nearby charge instability. To reveal the relationship between superconductivity and charge instability, we have investigated the charge ordered state by ^{13}C NMR spectroscopy at high magnetic fields. Our results clearly show that the modulation in the local density of state (DOS) appears below 8.5 K, and the DOS modulation forms a non-trivial 3-fold pattern [2], rather than the simple 2-fold rich-poor pattern. As the theory predicted that the strong charge fluctuations are induced near the charge ordering with such non-trivial charge pattern [3], the superconducting state in the charge ordered state would be affected by the charge instability. We discuss the relationship between charge instability and superconductivity in β‘‘-type BEDT-TTF salt, comparing with our recent results on α-type salts.

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Superconductivity in λ-(BEDT-STF)$_2$GaCl$_4$ under pressure

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The organic salt λ-(BEDT-STF)$_2$GaCl$_4$ consisting of us-BEDT-STF, situated between λ-(ET)$_2$GaCl$_4$ and λ-(BETS)$_2$GaCl$_4$ in the pressure phase diagram, shows an insulating behavior at ambient pressure. The electrical resistivity was measured in λ-(BEDT-STF)$_2$GaCl$_4$ under the pressure up to 2.1 GPa. We observed the sharp drop of resistivity associated with the superconducting transition around 5 K at 1.22 GPa for the first time. We confirmed the superconductivity by the pair-breaking under the magnetic field. The superconducting transition temperature T_c increases with increasing pressure up to 1.35 GPa as shown in Fig. 1. It decreases with the pressure increase above 1.35 GPa and the superconductivity disappears at 2.1 GPa. These behaviors indicate that λ-(BEDT-STF)$_2$GaCl$_4$ is definitely located at lower pressure region than λ-(BETS)$_2$GaCl$_4$, which has the superconducting phase at ambient pressure. The maximum T_c=5.95 K a little lower than 9 K in λ-(BETS)$_2$GaCl$_4$ is probably due to the randomness introduced by unsymmetrical donor us-BEDT-STF. We also investigated the magnetic property of λ-(BEDT-STF)$_2$GaCl$_4$ at ambient pressure by the static susceptibility and the ^{1}H NMR measurements. Although there is an increase of T_1^{-1} in ^{1}H NMR towards the low temperature, the magnetic order was not found down to 2 K. The observed spin susceptibility with a broad peak around 65 K is well fitted by that of the spin 1/2 two-dimensional Heisenberg AF in the triangular lattice with exchange coupling J=165 K. These suggest that the frustration suppresses the AF order, which is stabilized at 13 K in λ-(ET)$_2$GaCl$_4$ with lower chemical pressure, in λ-(BEDT-STF)$_2$GaCl$_4$. On the other hand, the randomness may cause the suppression of the AF phase. In any case, it is interesting that the neighboring phase of the superconductivity in λ-(BETS)$_2$GaCl$_4$ is not the AF phase in contrast to κ-(ET)$_2$X system.

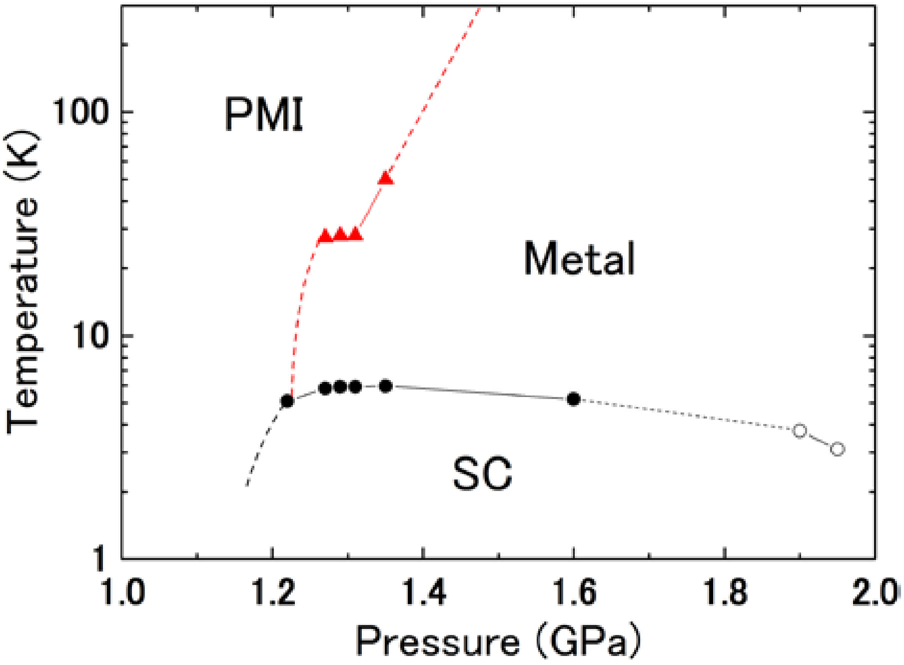

Figure 1: P-T phase diagram. Open and solid circle represent the onset Tc obtained from sample A and B

PI-30

ESR investigation of spin-dynamics in low-dimensional organic conductors $(TMTTF)_2X$

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$(TMTTF)_2X$ salts have been established as typical materials with low-dimensional electronic system. Noting that spin-Peierls, commensurate spin density wave (C-SDW), incommensurate spin density wave (IC-SDW), and superconducting phases are realized by applying appropriate pressures in the same $(TMTTF)_2X$ salts. Moreover, charge ordering phase exists in the paramagnetic region. The motivation of the present study is to clarify the competed electronic phases in $(TMTTF)_2X$ salts. Previously, we examined the spin dynamics in the vicinity of the spin-gap phase transition of the organic conductor $(TMTTF)_2PF_6$ by pulsed-ESR spectroscopy, comparing conventional spin-Peierls phase in $MEM(TCNQ)_2$ [1]. An anomaly in the relaxation time was observed in this intermediate region between the ground state and the high-temperature phase and was associated with the charge re-orientation originating in the transverse magnetic interaction. Considering the inter-chain interactions, the transverse magnetic interaction (triangular frustration network) plays an important role of the spin-gap phase transition in $(TMTTF)_2PF_6$. Recently Kawamoto and coworker found anomalous ^{13}C-NMR spectra change in $(TMTTF)_2Br$ at around 4.2 K. Since the antiferromagnetic state of $(TMTTF)_2Br$ was clarified as commensurate [2], IC-C sub-phase transition seems unlikely. Hence we also investigated antiferromagnetic resonance (AFMR) of $(TMTTF)_2Br$ dwon to 1.5 K. Curious temperature dependence of AFMR was observed below the Neel temperature. Possible sub-phases in the antiferromagnetic state are discussed.

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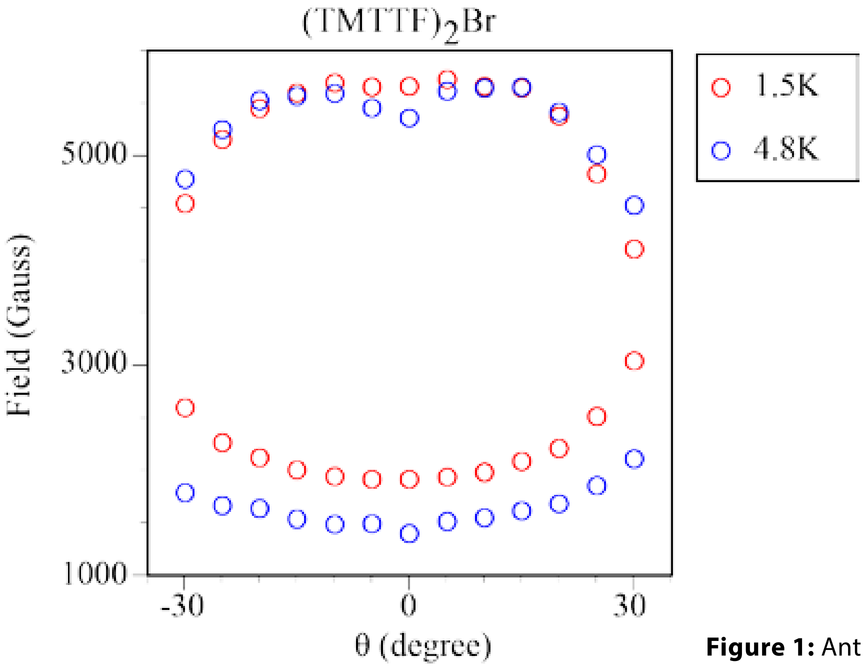

Figure 1: Antiferromagnetic Resonance in $(TMTTF)_2Br$.

PI-31

Comprehensive resistivity and Hall effect studies on quasi-one-dimensional $(TMTTF)_2X$ salts ($X=BF_4$, ReO_4, AsF_6, SbF_6)

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Upon cooling the highly anisotropic organic conductors $(TMTTF)_2X$ undergo transitions to various ordered states in the charge and spin degrees of freedom and therefore turn out to be an excellent choice for exploring the phenomena of low-dimensional physics. In order to study the transport mechanism of these quasi-one-dimensional (1D) organic salts, the temperature-dependent Hall effect of $(TMTTF)_2X$ ($X=BF_4$, ReO_4, AsF_6, SbF_6) has been measured by an AC compensation technique with current injection along the high-conducting a-axis. At elevated temperatures the Hall coefficient R_H of all investigated compounds seems to be constant, positive and somehow enhanced compared to the calculated value for one hole per unit cell. Beyond the charge order phase transition the Hall coefficient follows similar exponential laws as the resistivity data. This indicates the behavior of a semiconductor, where the change in $\rho(T)$ mainly originates from the change in the number of carriers. Here we discuss how the Hall coefficient is influenced by anisotropy, localization effects, anion symmetry and charge- as well as anion-ordering phenomena at low temperatures. We compare the results to resistivity studies and theoretical expectations.

PI-32

Monotonic growth and other new features of interlayer magnetoresistance in strongly anisotropic quasi-2D compounds

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The organic metals, usually, have layered molecular structure, which results in a strong anisotropy. Magnetic field is commonly used a powerful tool to study such electronic structure. Interlayer magnetoresistance in strongly anisotropic quasi-2D metals, where the interlayer hopping time is comparable to the cyclotron period, has many qualitatively new features, not described by the standard 3D theory. The classification of different regimes of interlayer magnetotransport in quasi-2D metals is governed by the ratios of three energy parameters: (i) Landau-level separation $\hbar\omega_c$, (ii) interlayer transfer integral t_z and (iii) the level broadening Γ_0 due to disorder. In my talk I briefly summarize the recent theoretical results on interlayer magnetoresistance in various regimes, which strongly differ from the predictions of the standard theory. In particular, in high magnetic field B_z, when the Landau levels become separated, the monotonic part of interlayer magnetoresistance $R_{zz}(B_z)$ strongly depends on the on the out-of-plane component of magnetic field B_z without saturation (square-root dependence), contrary to the prediction of no longitudinal magnetoresistance from the standard theory in the tau-approximation. This effect changes the angular dependence of magnetoresistance. The Landau-level shape in the very anisotropic quasi-2D metals is not Loretzian, which affects the amplitude of angular magnetoresistance oscillations. In some special cases, when the diffusion of electrons along the conducting layers is slow, the Coulomb e-e interaction may further increase the interlayer magnetoresistance.

[1] P.D. Grigoriev, T.I. Mogilyuk, Phys. Rev. B 90, 115138 (2014); A.D. Grigoriev and P.D. Grigoriev, Low Temp. Phys. 40(4), 367 (2014); P.D. Grigoriev, to be published.
[2] P.D. Grigoriev, Phys. Rev. B 88, 054415 (2013).
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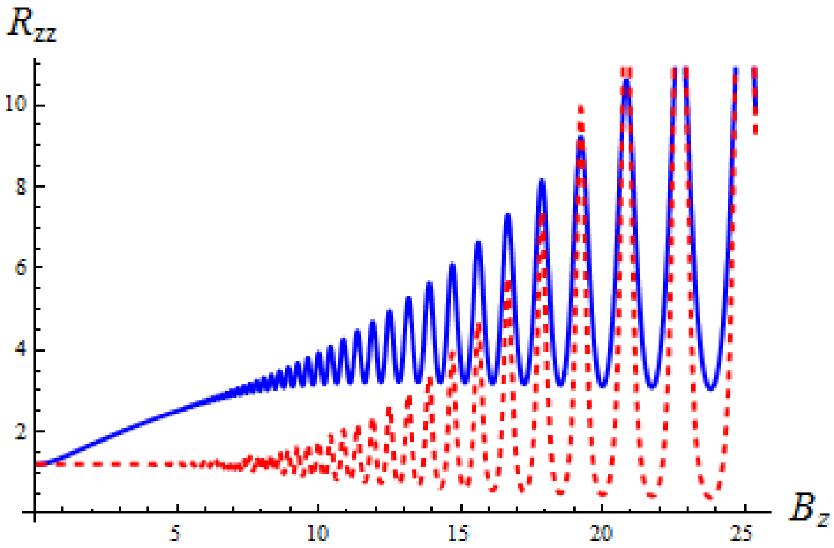

Magnetoresistance: The longitudinal interlayer magnetoresistance in quasi-2D metals, predicted by the standard theory (red dashed line) and by the new formula (solid blue line).

PI-33

Resonant Raman scattering in ET salts with Re_2X_8 (X=Cl, Br) anions

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The ET (or BEDT-TTF) is a well known electron donor molecule which forms a lot of the charge transfer (CT) salts with various anions. Among the variety of anions used for synthesis of CT salts there are only a few containing metal-metal bond [1,2]. Recently, Reinheimer et. al. synthesized a series of the CT salts with the metal-metal bonded anions [3]. We followed that trend and decide to take a look on the spectral properties of such systems. The two ET salts of Re_2X_8 (X=Cl, Br) anions have been syntesized and characterized using Raman scattering technique. Several excitation wavelengths were used to obtain Raman spectra and resonant Raman spectra. The spectra consist of the bands related to vibrations of the ET and the anion. In both salts the ET is carrying the +1 charge what is confirmed by the position of the $\nu_2(A_g)$ and $\nu_3(A_g)$ modes at 1452 and 1405 cm^{-1}. The spectra obtained with Ar ion laser (488 and 514 nm) are dominated by ET vibrations while the spectrum obtained with He-Ne laser (633 nm) is clearly dominated by the anion vibrations. The intensities of anion bands are enhanced due to resonance effect; moreover, combinations and overtones are observed. The position of bands related to the stretching of Re-Re bond at 275 cm^{-1} confirms the quadruple nature of the metal-metal bond in the anion. The resonant effect is observed for both salts, however the spectra of $ET_2Re_2Br_8$ shows smaller number of vibrational features.

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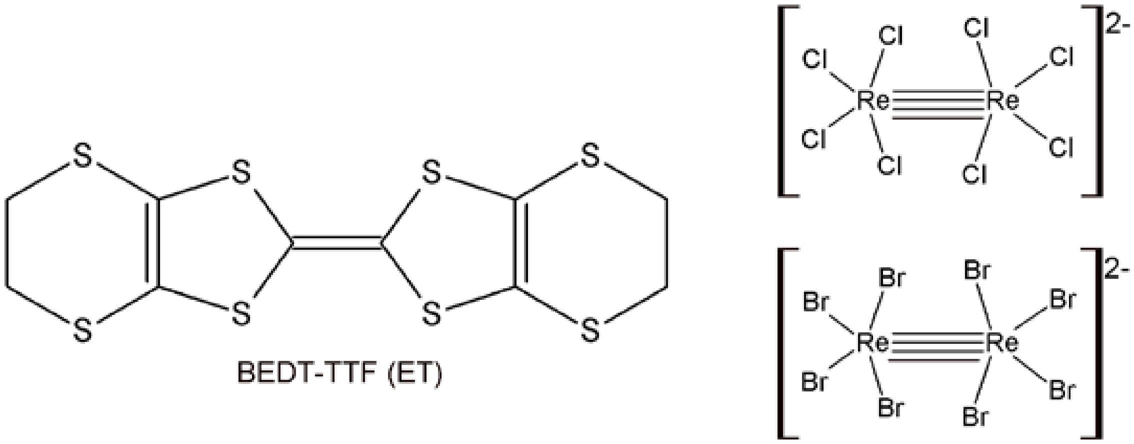

The components of salts.: The molecular structure of the ET and anions.

PI-34

In-Plane anisotropy of flux-flow resistivity in layered organic superconductor β“-$(ET)_2SF_5CH_2CF_2SO_3$

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[3]*Argonne National Laboratory, Materials Science Division, United States*

We report the in-plane anisotropy of the upper critical field (H_{c2}) and the flux-flow resistivity (FFR) for a layered organic superconductor β“-$(ET)_2SF_5CH_2CF_2SO_3$ with the incoherent nature of the interlayer transport[1]. The in-plane angular dependence of H_{c2} shows a fourfold oscillation with maxima in the directions around H parallel to the b and a axes. This result is compatible with a d_{x2-y2} order parameter. The determined nodal structure is consistent with theoretical predictions of superconductivity mediated by charge fluctuations[2]. Figure 1 shows the dependence of the FFR on in-plane field orientation in the non-Ohmic regime. As seen in Fig. 1, the FFR shows a fourfold symmetry with cusp-like minima in the directions around H parallel to b and a axes, which is very similar to the FFR for κ-$(ET)_2Cu(NCS)_2$ with d-wave pairing symmetry[3]. From these results, we claim that the vortex dynamics is strongly affected by the SC gap structure for highly two-dimensional superconductors with d-wave pairing symmetry.

[1] J. Wosnitza, J. Hagel, J. S. Qualls, J. S. Brooks, E. Balthes, D. Schweitaer, J. A. Schlueter, U. Geiser, J. Mohtasham, R. W. Winter, and G. L. Gard, Phys. Rev. B 65, 180506(R) (2000).
[2] J. Merino and R. H. Mckenzie, Phys. Rev. Lett. 87, 237002 (2001).
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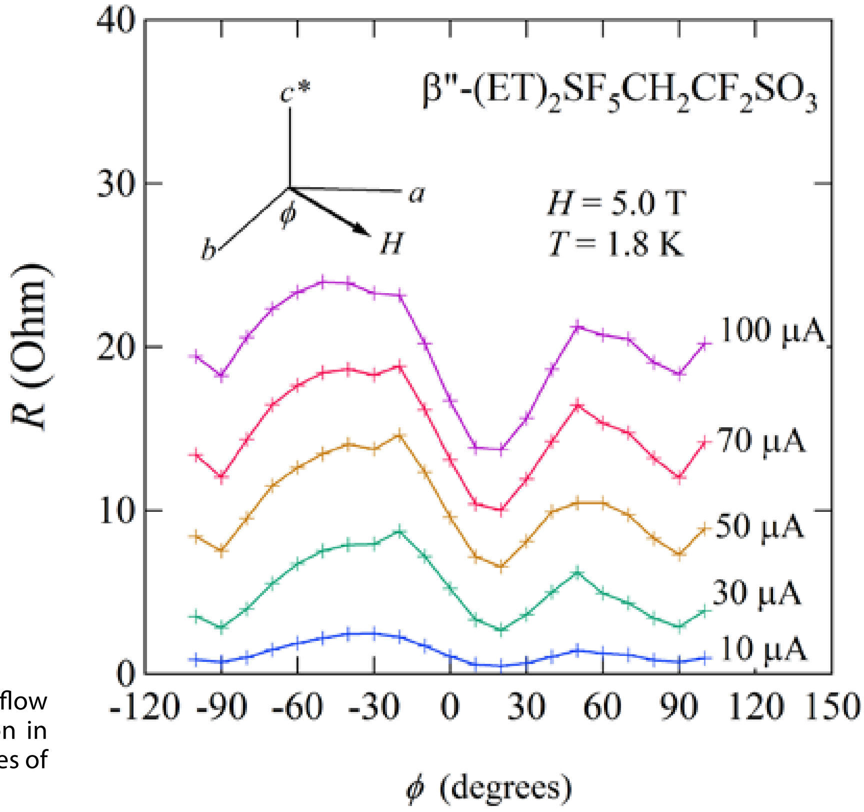

Figure 1: The dependence of the flux-flow resistance on in-plane field orientation in the non-Ohmic regime for several values of current.

PI-35

Conductivity and thermopower of organic conductors (BTBT)$_2$X (X: PF$_6$,AsF$_6$,SbF$_6$,TaF$_6$)

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Organic semiconductor [1]benzothieno[3,2-b][1]benzothiophene (BTBT) (inset figure) is known to show high performance in an organic field effect transistor. Recently, a (BTBT)$_2$PF$_6$ charge transfer complex using BTBT has been reported by T. Kadoya et al [1]. This complex shows high conductivity up to 1500 S/cm, though the calculated band width is as small as 0.35 eV. Usually, Seebeck coefficient is inversely proportion to the band width. Therefore this complex is expected to be a good thermoelectric material because of the high conductivity and the small band width. Here, we report new BTBT charge transfer complexes : (BTBT)$_2$AsF$_6$, (BTBT)$_2$SbF$_6$, and (BTBT)$_2$TaF$_6$. We measured the resistivity, Seebeck coefficient, and ESR. The obtained crystals are black rods with tetragonal symmetry composed of one-dimensional uniform stacking along the c axis. The resistivity is metal-like around room temperature and becomes insulating accompanied by a non-continuous resistivity jump around 150 K. When coated with Apiezon N grease, resistivity jump is suppressed, and the resistivity becomes metal-like down to 60 K, below which it becomes insulating. However, the insulating phase is paramagnetic. The maximum conductivity of (BTBT)$_2$AsF$_6$, which is larger than (BTBT)$_2$PF$_6$, is as large as $\sigma = 4100$ S/cm. The Seebeck coefficient is metal-like without coated (Figure). The room temperature value is about $S = 15$ mV/K, from which the band width is estimated to be about 1.4 eV. The power factor $PF = \sigma S^2$, which is an index of thermoelectric efficiency, is 7.7×10^{-5} (W/K^{2}m). This value is very large among organic materials.

[1] T. Kadoya et al., Phys. Chem. Chem. Phys., 15, 17818 (2013).

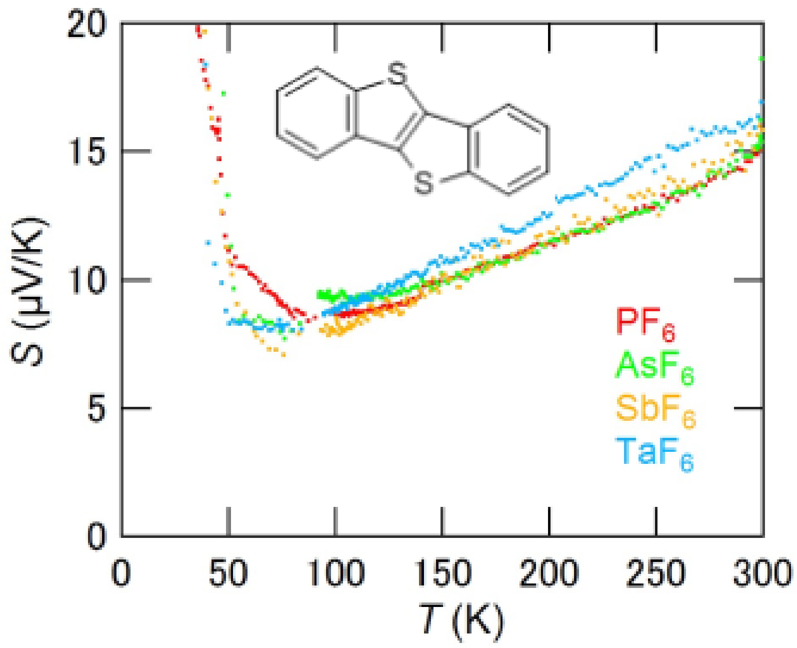

Figure: Temperature dependence of thermoelectric power and a BTBT molecule (inset)

PI-36

Magnetocaloric effect in antiferromagnetic insulating phase of λ-(BETS)$_2$FeCl$_4$

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[2]*National Institute for Materials Science, Tsukuba, Japan;*
[3]*Toho University, Department of Physics, Funabashi, Japan;*
[4]*RIKEN, Condensed Molecular Materials Lab., Wako, Japan;*
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A two dimensional organic conductor λ-(BETS)$_2$FeCl$_4$ is known to show a metal-insulator transition at about 8 K, associated with an antiferromagnetic (AF) order. The antiferromagnetic insulating (AFI) phase is broken by a magnetic field of about 10 T, and then the paramagnetic metallic (PM) phase is recovered. [1] In these phase transitions, the strong exchange interaction J between the π spin with S = 1/2 and 3d spins with S = 5/2, so-called π-d interaction, plays a crucial role. The mechanism of the AFI-PM transition has been extensively studied so far but it is still controversial. Recent specific heat studies[2] show the AF order of the π spins in the AFI phase, which contradicts the previous interpretation, the 3d spin order. In order to further investigate the AFI-PM transition, we have performed magnetocaloric measurements. In this experiment, the sample temperature is measured in a semi-adiabatic condition as a function of field. The magnetocaloric effect, leading to the sample temperature change ΔT, is quite sensitive to the entropy (S) change dS/dH. In λ-(BETS)$_2$FeCl$_4$, ΔT arises mainly from the entropy of the localized spin state. Figure 1 presents ΔT as a function of field (H//c) at some different bath temperatures T_0. As the field increases for T_0 = 1.3 K, ΔT shows a dip at about 1 T, a broad minimum at about 5 T and then a sharp dip at about 10 T (AFI-PM transition). Above 10 T (PM phase), ΔT gradually decreases with increasing field, which corresponds to the entropy decrease of the paramagnetic 3d spins. The broad minimum and sharp dip depend strongly on the bath temperature whereas the dip at about 1 T seems almost temperature independent. The details will be discussed in terms of the entropy of the spin states and the strong π-d interaction.

[1] H. Kobayashi et al., J. Am. Chem. Soc., 118, 368-377 (1996)
[2] H. Akiba et al., J. Phys. Soc. Jpn., 78, 033601 (2009)

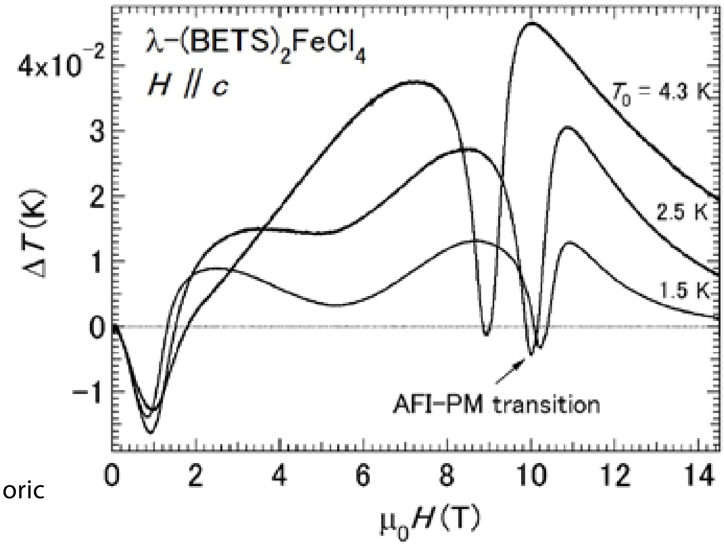

Figure 1: Magnetic field dependence of magnetocaloric effect for H//c axis.

PI-37

The influence of Coulomb blockade on the interlayer electron transport in strong magnetic field

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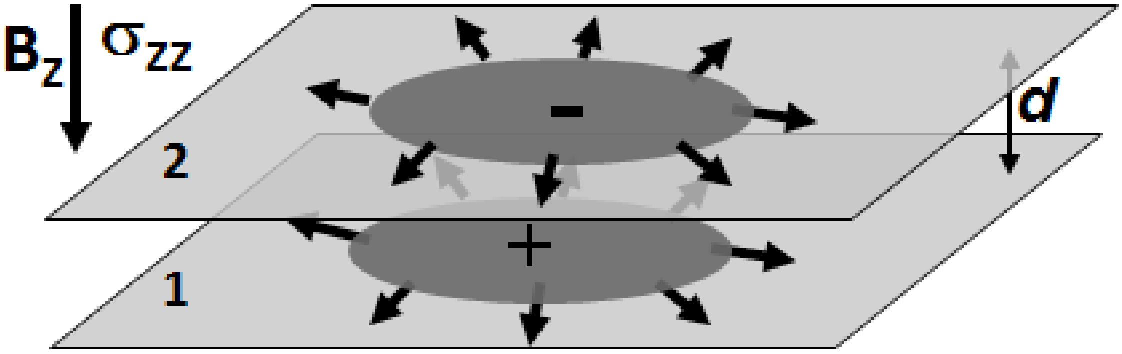
The interlayer conductivity in organic metals is, usually, determined by the electron tunneling between adjacent molecular conducting layers. The electron-electron interaction may suppress such tunneling interlayer transport due to the Coulomb blockade effects. The Coulomb blockade effect is strong if the charge dissipation along the conducting layers is slow, so that the electron hopping between two adjacent layers 1 and 2 costs an additional Coulomb energy between the hopped electron on layer 1 and a hole (non-dissipated polaron) on layer 2 for a long enough time. The perpendicular-to-layers external magnetic field reduces the metallic in-plane conductivity, which increases the Coulomb blockade effects and may lead to a strong longitudinal interlayer magnetoresistance $R_{zz}(B_z)$. Without the charge diffusion (in the case of electron localization) this would give an exponential magnetoresistance $R_{zz}(B_z)$, but the finite in-plane charge diffusion leads to a power-law dependence $R_{zz}(B_z)$. Using the calculation by the path-integral method in the quasi-classical approximation, developed in [1], we analyze the influence of the Coulomb blockade effects on the interlayer magnetoresistance in some organic metals, e.g. in b-(BEDT-TTF)$_2$SF$_5$CH$_2$CF$_2$SO$_3$, and in GaAs/AlGaAs heterostructures, where the strong longitudinal interlayer magnetoresistance has been observed in many experiments.

[1] L.S. Levitov and A.V. Shytov, JETP Lett. 66, 214 (1997)



Scheme of two layers: The schematic view of the charge distributions (dark-gray areas) of polaron (lower layer) and of the electron (upper layer) after an electron jump to the adjacent layer. The arrows show the charge diffusion along the layers.

PI-38

Charge stripes in graphene sheets of CaC_6 intercalated compound

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The recent experimental discovery of charge stripes in graphene sheets within intercalated CaC_6 compound [1] has drawn a lot of attention in physical community. Most importantly, the inability to explain the origin of such charge modulation via standard nesting mechanism implies the necessity of introduction of new concepts in understanding the physics of density waves. We propose a model in which we utilize pseudomagnetic fields, induced by lattice deformation, that influence electron dynamics via magnetic breakdown mechanism, to explain the onset of the density modulation. The physical properties of graphite is significantly changed in CaC_6 compound that is formed by intercalation of graphite with Ca atoms that form a hexagonal superlattice between carbon sheets. Among other changes, they dope graphene with ~0.2 electrons per carbon atom, resulting in triangularly shaped Fermi surfaces. The observed charge density wave appears along the underlying Ca^{2+} ion line and has period in perpendicular direction equal to three times that of the Ca superlattice. Taking all counted, the nesting mechanism is ruled out. However, the observed density wave periodicity relates Fermi surfaces exactly to slightly overlaping position. Earlier, we proposed a mechanism by which the density wave can be induced by magnetic breakdown, that lowers total energy of the system, in such configuration due to an external magnetic field [2]. Here, a slight, spontaneous, inplane deformation of carbon lattice induces a perpendicular pseudomagnetic field that can be large (few hundred Tesla). Magnetic breakdown, induced by that field, connects closed, slightly overlapping electron orbits [3], thus delocalizing an electron and lowering its energy. The final fine tuning of density wave position is a result of energy balance between minimum of energy due to magnetic breakdown band configuration and electronic interaction between density wave and the underlaying Ca^{2+} superlattice.

[1] K. C. Rahnejat et al., Nature Commun., 2:558 (2011); Howard et al., SPIE Newsroom 10.1117/2.1201201.004112.
[2] A. M. Kadigrobov, A. Bjeliš and D. Radić, Phys. Rev. Lett. 100 (2008) 206402; A. M. Kadigrobov, A. Bjeliš and D. Radić, Eur. Phys. J. B: condensed matter physics 86 (2013) 276.
[3] A. M. Kadigrobov, D. Radić, and A. Bjeliš, Physica B: Condensed matter 460 (2015) 248.

PI-39

Microscopic study of donor-acceptor type one dimensional charge transfer complex, HMTSF-TCNQ

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[4]*RIKEN, Wako, Japan*

Donor-acceptor-type charge transfer complex, HMTSF-TCNQ is a one-dimensional conductor in which both HMTSF donor and TCNQ acceptor stacks are considered to contribute to the electric properties. At ambient pressure, it is reported that the system behaves metallic below room temperature and undergoes a metal-insulator transition due to CDW formation at about 30 K. Applying pressure suppresses the transition. At a high pressure of 1.1 GPa, unconventional magneto-transport behaviors have been observed. The electronic state under pressure/high magnetic field is expected as a field-induced CDW (FI-CDW) state, caused by a similar scenario as the FI-SDW in TMTSF2X family. We have started to study this material (at ambient pressure) from microscopic points of view. We performed Se NMR measurements to investigate the electronic properties on HMTSF site, and found temperature independent NMR shift and exponentially decreases of relaxation rate with decreasing temperature below 200K. These results strongly suggest nonmagnetic natures of the HMTSF stack in this temperature region. There were no anomaly around 30 K which is reported as the CDW transition temperature. We also performed X-band EPR measurements using the same crystal to detect electrons on the TCNQ stack. No significant EPR signal was observed in the whole range of measured temperature. The idea that ``only TCNQ stack contribute to the conduction" was ruled out. The observed NMR/ESR results seem inconsistent with the transport properties at least at ambient pressure. Further investigations to clarify the electronic structure microscopically, such as 1H/13C-NMR measurements, applying pressure, magnetic susceptibility measurements, etc. are underway.

PI-40

Dynamically enhanced two-dimensionality and emerging spin-liquid ground state in a molecular rotor system (EDT-TTF-CONH$_2$)$_2$BABCO

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The sensitivity of electronic and magnetic properties of organic molecular conductors to modifications of their crystal structures is responsible to the great complexity of their phase diagram. Details of the precise interactions between the organic slabs and the anion layers are of great importance. Materials where these interactions can be dynamically modulated through anion's movements open a new avenue to study the phase diagram. Here we report on multi-frequency electron spin resonance (ESR) spectroscopy in 0-420 GHz frequency range complemented by electronic transport measurements on (EDT-TTF-CONH$_2$)$_{2+}$[BABCO$^-$], a conducting molecular system where slabs of conducting organic radical cations of (EDT-TTF-CONH2)2+ alternate along c with the BABCO– molecular rotors [1]. (EDT-TTF-CONH$_2$)^{2}BABCO undergoes a strongly 2D metal to insulator transition at T_{MI}=50 K. The metallic phase is characterized by linearly decreasing Seebeck coefficient, indicative to a metallic, electron-like conduction band with ε_F=0.2 eV Fermi energy. ESR confirms the Pauly-like temperature independent spin susceptibility. However, the conductivity is extremely 2D. The anisotropy exceeds 105. Below T_{MI} as the insulating phase develops not only the charge but also the spin dynamics changes character. Strong J_{AF}=30 K antiferromagnetic correlations appear but no magnetic transition occurs down to at least 1.2 K, pointing towards a spin-liquid ground state. The spin relaxation time shows unconventional temperature and magnetic field dependence. We attribute these remarkable behaviors to the extreme two-dimensionality of the system. The coherence between the (EDT-TTF-CONH$_2$) slabs is destroyed by quantum dissipation introduced by the Brownian rotation of the BABCO– molecular rotors.

[1] C. Lemouchi et al. JACS 2012, 134, 7880-7891

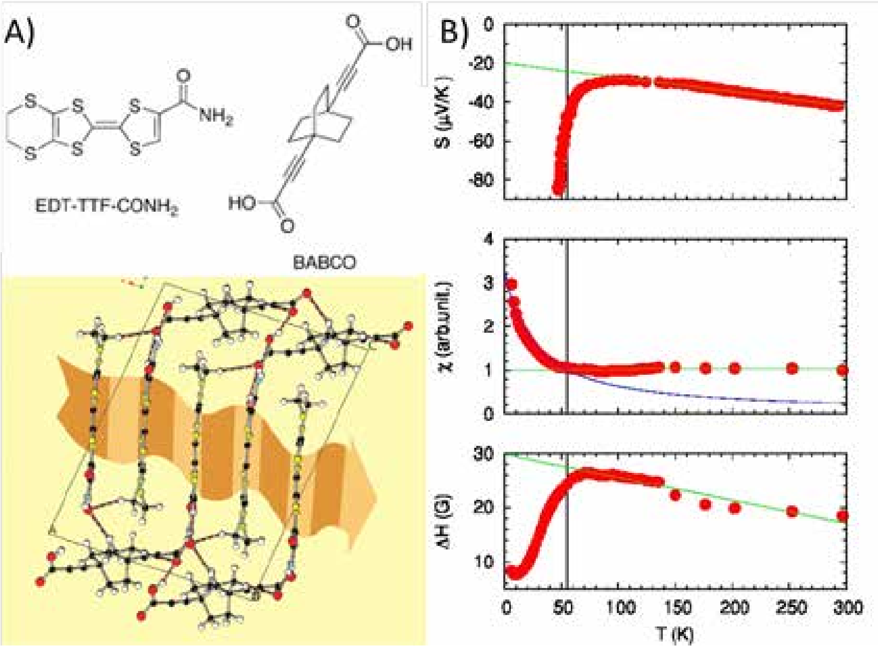

Figure 1: A) Structure of EDT-TTF-CONH$_2$, BABCO and (EDT-TTF-CONH$_2$)$^{2+}$[BABCO$^-$]. [1] B) Temperature depen-dence of the Seebeck coefficient (top), spin susceptibili-ty (middle) and ESR linewidth (bottom.)

Correlation between electronic and phononic excitations governing the nonlinear conduction in DCNQI$_2$Cu single crystals

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Nonlinear conduction has been reported in low-dimensional organic conductors for a variety of materials with different ground states at low temperatures. Some experimental features hint at collective excitations like solitons[1] being responsible for the observed phenomena. On the other hand, Mori et al. were able to simulate the nonlinear current-voltage characteristics of different organic charge-transfer salts by means of a general electrothermal model considering the electrical heating of distinct electronic or phononic subsystems of a material[2]. By analysis of the effective specific heat utilized in the model, an identification of the charge-carrying excitations governing the nonlinear conduction is possible. In this contribution we present a comprehensive analysis of the transient resistive response of deuterated DCNQI$_2$Cu single crystals in fields up to 3600 V/cm and on time-scales between 100µs and 1s applying the electrothermal model. We have been able to determine a temperature-dependent effective specific heat below 75K and found it to be reduced by about 40 J/(mole K) compared to steady-state literature values[3]. The magnitude and temperature-dependence can be explained by neglecting low-energy phonon excitations and modeling the data by an effective Einstein model based on a mean phonon energy of 19meV. Hence, we identify optical phonon degrees of freedom to govern the effective specific heat providing evidence for a charge-carrying excitation efficiently interacting with optical phonons, such as e.g. solitons[4]. This interpretation is supported by the coincidence between the average phonon energy and the observed activation energy of (17±2)meV in the low-field conductivity regime. In general, our measurements bridge the gap between the generic electrothermal model and the microscopic origin of the field-dependent conductivity and may also be applicable in the context of other low-dimensional conductors.

Financial support by the DFG (project PF385/6-1) and the 7th Framework Program (H2ESOT) of the European Commission is gratefully acknowledged. We also like to thank Prof. Markus Schwoerer and Prof. Martin Dressel for fruitful discussions.

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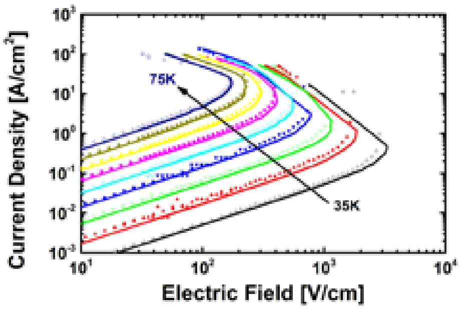

Nonlinear Conduction:
Nonlinear Current-Voltage Characteristics of (DCNQI)2Cu between 35K and 75K together with simulations applying an electrothermal

PI-42

The potential of quasi-one-dimensional organic metals for thermoelectric applications

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By their exceptional electronic and thermal properties organic conductors are interesting candidates for near-ambient temperature thermoelectrics in which conventional semiconductors and metals are limited due to their inherent properties. The dimensionless figure of merit $ZT=S^2\sigma T/\kappa$, depending on the Seebeck coefficient S, the electrical conductivity σ and the thermal conductivity κ, determines the Carnot limit of a thermoelectric device. Since high electrical and low thermal conductivities impose fundamental demands on materials suited for thermoelectric applications, the predicted violation of the Wiedemann-Franz law in quasi-one-dimensional organic charge transfer complexes1 gives rise to consider this class in more detail. In this contribution, we present first comprehensive temperature-dependent studies of Seebeck coefficient, electrical and thermal conductivity on organic p- and n-type single crystals like tetrathiotetracene-iodide (TTT_2I_3) and copper radical anion salts of dicyanoquinonediimine ($DCNQI_2Cu$). In order to estimate all relevant physical quantities contributing to ZT on just one sample an electro-thermal measurement setup has been installed. We obtained maximum power factors $P= S^2\sigma$ of $1.4\cdot10^{-4}$W/(mK^2) for TTT_2I_3 and $2.5\cdot10^{-5}$W/(mK^2) for $DCNQI_2Cu$ being smaller by about one or two orders of magnitude compared to conventional thermoelectric materials like Bi_2Te_3. To allow for potentially high ZT values in low-dimensional organic metals, a detailed understanding of the thermal conductivity is necessary for which we present preliminary results including an investigation of the Wiedemann-Franz law.

Financial support by the EU FP7 project H2ESOT is gratefully acknowledged.

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Superconducting properties and pressure effect on In doped topological crystalline insulator SnTe

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Abstract – We are reporting the superconducting properties of superconductor $Sn_{0.5}In_{0.5}Te$, derived from topological insulator SnTe. Silvery shiny crystals were grown using modified Bridgman method in a vertical tube furnace. XRD on powdered sample and Rietveld refinement has confirmed the phase purity of the sample. Superconducting transition temperature T_c at 4.5 K was achieved in the resistivity measurement done from 1.6-300K temperature range. Magnetic measurement has confirmed bulk superconductivity at same T_c. Upper critical field (by WHH formula) and Coherence length has been estimated be 1.6 T and 143.5 Å. Majority of positive charge carriers in the crystal has been found by Seebeck and Hall coefficient measurement. The negative coefficients of T_c suppression of -0.66 K/GPa has been found on the application of the pressure upto 3 GPa.

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PI-44

Negative Hall state in the FISDW phase of $(TMTSF)_2ReO_4$

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Quasi-One-Dimensional organic conductors $(TMTSF)_2X$ shows a field-induced spin-density-wave (FISDW) phase. The FISDW phase has a cascade of subphases characterized by the quantized Hall resistance $\rho_{xy} = h/2e^2N$. In the PF_6 salt under 0.7 GPa, the sequence of N shows N = 0, 1, 2, -2, 3, 4, 5, -4, 6, … with decreasing field[1]. This can be explained as the competition between the positive integer and negative even N phases[2][3]. On the other hand, in the ClO_4 salt at ambient pressure, the sequence shows N = 0, 1, 3,-1, 5, … which means the suppression of the positive even N phases[4]. To explain the difference between the PF_6 and ClO_4 salts, attention has been focused on the superlattice along b direction due to the anion ordering (AO). However, the origin of the difference is still in question. In order to solve this problem, we measured the magneto and Hall resistance of the ReO_4 salt which also has the AO above 1.0 GPa[5]. The size of the AO gap, which is determined from the rapid oscillation in the amgnetoresistance, is almost same to the one in the ClO_4 salt. We found that the sequence is N = 2, -2, 4, -4, -6, … below 17 T. There is no suppression of even N phases and the negative even N phases appear in the wide magnetic field range. In the presentation, we will discuss the origin of the FISDW phase diagram with AO. The difference between the ReO_4 and PF_6 salts suggests that the AO suppresses positive integer N phases. As a result, negative even N phases become wider. The difference between the ReO_4 and ClO_4 salts suggests the significance of difference between the hydrostatic and chemical pressures. We deduced that the sequence of N is determined by the nesting vectors. The sequence in the PF_6 salt and the ReO_4 salt can be explained by a standard nesting vector $Q_0=(2k_F,\pi/b)$. On the other hand, one in the ClO_4 salt can be explained by a peculiar nesting vector $Q\pm=(2k_F\pm\delta,\pi/2b)$.

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Optical study of the metal-insulator phase transition in chiral molecular conductor [(S,S)-DM-EDT-TTF]$_2$ClO$_4$

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Chiral charge transfer salts [(S,S)-DM-EDT-TTF]$_2$ClO$_4$ and [(R,R)-DM-EDT-TTF]$_2$ClO$_4$ are newly discovered materials that display an electrical magnetochiral anisotropy effect [1]. These molecular conductors are based on the chiral dimethylethylenedithio-tetrathiafulvalene (DM-EDT-TTF) donor molecules [2] (Fig. 1) and crystallize in the enantiomorphic hexagonal space groups P6222 ([(S,S)-DM-EDT-TTF]$_2$ClO$_4$) and P6$_4$22 ([(R,R)-DM-EDT-TTF]$_2$ClO$_4$). Band structure calculations suggest one-dimensional metallic conductivity, together with possible CDW or SDW instabilities at low temperature. Metal-insulator phase transition (MI) is observed at 40 K in both salts [1]. Here, temperature-dependent infrared reflectance spectra of [(S,S)-DM-EDT-TTF]$_2$ClO$_4$, are studied in order to provide more information about the origin of the MI transition in this material. Our results display no polarization dependence in the conducting plane. Both electronic and vibrational features display significant changes with lowering the temperature. Signatures of the lattice distortion related to MI are found at 40 K, suggesting CDW origin of the transition.

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This work was supported by the Polish National Science Centre (Decision No. DEC-2012/04/M/ST3/00774).

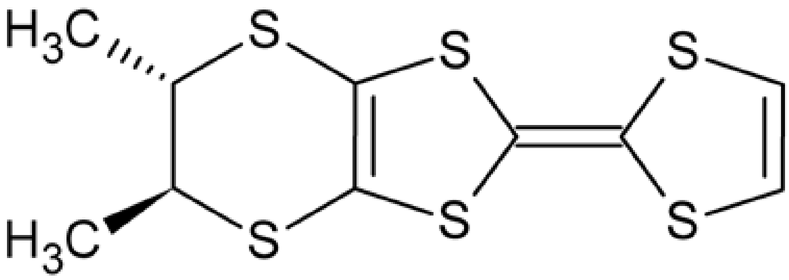

Figure 1: (S,S)-DM-EDT-TTF donor molecule

PI-46

Metallic transport near a quantum critical point in organic superconductors from a renormalized Boltzmann theory

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The electrical and thermal transport properties of the normal state of quasi-1D superconductors like Bechgaard salts are investigated by combining the linearized Boltzmann equation and the renormalisation group (RG) method. The collision integral operator is calculated using the Umklapp scattering amplitudes obtained by the RG method yielding the electrical resistivity (ρ) and Seebeck coefficient (S). The power law dependence, $\rho \sim T^{\alpha}$, for resistivity is obtained by changing the antinesting parameter t'_b simulating the pressure distance from the quantum critical point (QCP) between spin-density-wave (SDW) and d-wave SC (SCd) in the phase diagram. The resistivity evolves from a linear component ($\alpha \approx 1$) at the QCP towards a Fermi liquid component ($\alpha \approx 2$) with increasing t'_b, which confirms an extended region of quantum criticality as a result of interference between SCd and SDW causing an anomalous growth of Umklapp scattering. Its anisotropy is also tied to the transverse momentum k_{b-} dependence of hot/cold scattering regions along the Fermi surface. Similar calculations for the Seebeck coefficient show deviations from the usual linear temperature dependence and also a change of sign near a SDW instability.

PI-47

Resistivity and dielectric studies on κ-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl under He-gas pressure

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Charge-transfer salts of the κ-(BEDT-TTF)$_2$X family have been intensively studied over more than two decades with special focus lying on the Mott metal-insulator transition for pressurized X = Cu[N(CN)$_2$]Cl and the intriguing states evolving nearby such as superconductivity or multiferroicity. Compared to these systems, very little has been known about the title compound, first synthesized by Lyubovskii et al. [1]. This system, crystallizing in a monoclinic structure, shares the κ-type packing motif, implying magnetic frustration t'/t. It undergoes a metal-insulator transition around 30 K which has been assigned to a charge-ordering transition [2]. According to electronic band structure calculations by Jeschke et al., the ratio t'/t is larger as compared to the Mott insulator X = Cu[N(CN)$_2$]Cl, whereas the degree of dimerization is less strongly pronounced, placing this material closer to a quarter-filled situation. Here we report on measurements of the dc-resistivity both at ambient and finite He-gas pressure on various single crystals of κ-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl. These studies are complemented by preliminary measurements of the dielectric constant on one crystal at ambient pressure. The resistivity data reveal a well-pronounced and sharp metal-insulator transition which can be rapidly suppressed by the application of hydrostatic pressure. A pressure of P = 97 MPa is sufficient to stabilize the metallic state down to 2 K, the lowest temperature of our experiment. By increasing the pressure to P = 146 MPa, the resistivity further decreases but no indications of superconductivity could be observed. Interestingly, measurements of the dielectric constant uncover a relaxor-type ferroelectric anomaly in the insulating low-T state.

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PI-48

Origin of the glass-like structural dynamics in molecular metals κ-(BEDT-TTF)$_2$X: implications from fluctuation spectroscopy and *ab initio* calculations

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[2]*Argonne National Laboratory, Materials Science Division, United States*

We have studied the low-frequency dynamics of the charge carriers in different organic charge transfer salts κ-(BEDT-TTF)$_2$X with polymeric anions X = Cu[N(CN)$_2$]Cl, Cu[N(CN)$_2$]Br, Cu(NCS)$_2$ by using resistance noise spectroscopy. We aim to investigate the well-known structural, glass-like transition caused by the conformational degrees of freedom of the BEDT-TTF molecules' terminal ethylene groups. Although of fundamental importance for studies of the electronic ground-state properties [1], the phenomenology of the glassy dynamics is only scarcely investigated and its origin is not understood. Our earlier studies of fluctuation spectroscopy revealed a universal, pronounced maximum in the resistance noise power spectral density related to the glass transition [2]. The energy scale of the fluctuations can be identified with the activation energy of the glass-like structural dynamics as determined from thermodynamic and NMR measurements. Here we present a systematic study of the various compounds. For the first time for this class of ‚plastic crystals', we report a typical glassy property of the relaxation time, namely a Vogel-Fulcher-Tamman law, and are able to determine the degree of fragility of the glassy system. Supporting ab initio calculations provide an explanation for the origin of the glassy dynamics in certain systems in terms of the coupling between the ethylene endgroups' degrees of freedom and the anion layer.

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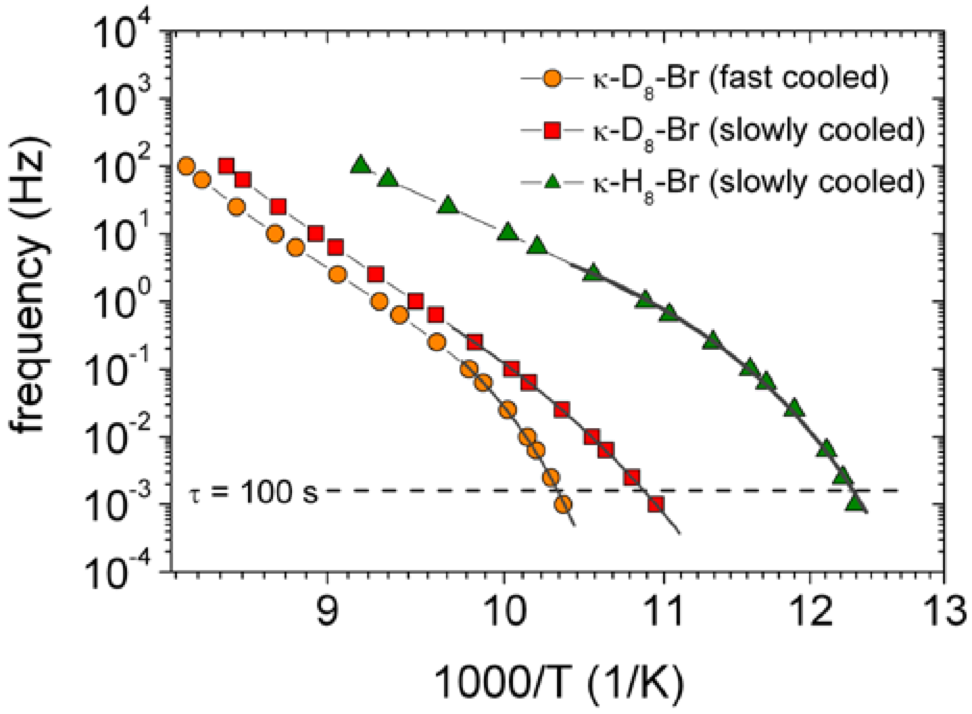

Figure: Arrhenius plot of the noise peak frequency for deuterated and non-deuterated k-(ET)$_2$Cu[N(CN)$_2$]Br showing a Vogel-Fulcher-Tammann behavior.

Magnetoresistance properties of κ-$(BETS)_2FeCl_4$: Revealing the phase diagram and mapping the Fermi surface

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[1]*Walther-Meißner-Institut, Garching, Germany;*
[2]*Technische Universität München, Physik-Department, Garching, Germany;*
[3]*Institute of Problems of Chemical Physics, Chernogolovka, Russia*

The bifunctional organic conductors $(BETS)_2$ FeX_4 (X = Cl or Br) are of high interest due to correlation effects and magnetic ordering coexisting with superconductivity. κ-$(BETS)_2FeCl_4$ contains magnetic moments, which show an antiferromagnetic (AFM) order below the Néel temperature TN ≈ 0.45 K [1]. Furthermore, this compound is reported to show superconductivity (SC) in the AFM state below $T_c \approx 0.17$ K. Our measurements of the interlayer resistance showed no clear sign of bulk SC despite a high crystal quality. On the other hand, the AFM transition is clearly resolved in the interlayer resistance due to reduced scattering in the AFM state. This provides a way to determine the phase boundaries with a magnetic field applied along different crystallographic axes. Thus we have obtained the temperature – magnetic field phase diagram of κ-$(BETS)_2FeCl_4$, showing some remarkable features. Further, angular magnetoresistance oscillations (AMRO) in the normal metallic state were studied at different fields and rotation axes. In the AMRO spectrum contributions of the different parts of the Fermi surface could be resolved and allowed a mapping of the Fermi surface, including classical and magnetic-breakdown orbits. The Shubnikov-de Haas (SdH) oscillations associated with those orbits were also investigated. The p-d exchange has been determined by tracking the angle-dependent position of nodes in the SdH oscillations originating from the field-dependent spin-zero effect [2]. Our results will be compared with the sister compound κ-$(BETS)_2FeBr_4$ in order to obtain information about the influence of halogen substitution on the physical properties.

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PI-50

Experimental setup for pressure dependent dielectric measurements on low-dimensional organic conductors

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[2]*Institut za fiziku, Zagreb, Croatia*

Low-dimensional organic conductors are well established as model systems for strongly correlated electron systems. The interplay of electronic, spin and lattice degrees of freedom together with changes in dimensionality result in a rich phase diagram, which may include antiferromagnetism, superconductivity, spin-density-wave ordering, spin-Peierls behavior and charge ordering. The position in the phase diagram can be tuned by external pressure, chemical pressure, via substituting the anion, and temperature. Commonly, the change in the dielectric properties associated with a phase transition is investigated by means of temperature dependent optical spectroscopy and electrical transport measurements. To extend the experimental access to the phase diagram, we establish a setup for electrical transport measurements up to 110MHz in dependence of both, temperature and pressure. Here, we present the overall setup and discuss the challenges when measuring dielectric properties on organic conductors under pressure. Additionally, we will present and discuss first results on the quasi-one-dimensional charge transfer salts $(TMTTF)_2X$ ($X=BF_4$, ClO_4) compounds at ambient pressure.

PI-51

Ordered flux-lines lattices in the high-fields mixed state of quasi-2D organic superconductors

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[2]*Technion-Israel Institute of Technology, Schulich Faculty of Chemistry, Haifa, Israel*

We have observed ordered flux-lines lattices in layered (quasi 2D) organic superconductors in the virtually unexplored high-fields low-temperatures region of their H-T phase diagram. Exploiting μSR line-shape spectroscopy and its great sensitivity to magnetic flux-lines order, [1,2] we have found clear indications of ordered 3D vortex lattices in the mixed states of the quasi 2D organic superconductors κ-$(ET)_2Cu[N(CN)_2]Br$ and κ-$(ET)_2Cu(NCS)_2$ (κ-CuNCS) at T = 20 mK, under magnetic fields as high as 4 T for the former compound and 3 T for the latter. Furthermore, even at T = 1 K we have observed in the former compound ordered vortex lattices at fields as high as 2 T. This fields range is 2 to 3 orders of magnitude higher than the characteristic field of the crossover from the 3D flux-lines lattice to a multilayer of uncorrelated 2D lattices of pancake vortices, predicted for these compounds by the continuum elastic theory of the vortex lattice. [3] It is also found that the experimentally observed sharp sign change of the μSR line-shape asymmetry (skewness) parameter, associated with the loss of flux-line order, is consistent with melting of pancake vortex lattices within the layers, [4] rather than with a 3D-2D crossover. These observations reflect a robustness of the 3D vortex lattice to thermal fluctuations of pancake vortices in the individual SC layers. We suggest that in the high magnetic field region of the phase diagram investigated, small concentrations of defects that pin flux lines perpendicular to the superconducting layers, effectively resist the breakup of the entire 3D vortex lattice by thermal fluctuations. The primary source of this flux-line pinning may be a consequence of the high rate at which the samples were precooled in our experiments when loaded into the dilution refrigerator.

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PI-52

The effect of X-ray irradiation on the T-p phase diagram of κ-$(ET)_2Cu[N(CN)_2]Cl$

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Charge-transfer salts of the κ-$(ET)_2X$ family are known for having a ground state which can be drastically altered by the application of pressure. Especially for X = $Cu[N(CN)_2]Cl$, the full s-shaped transition line between the Mott insulating and the metallic phase is accessible by applying pressures of no more than 30 MPa [1]. As a result, this salt has served as a model system for exploring the Mott transition and the anomalous states that form nearby. An aspect of considerable recent interest in this field of research is the interplay of strong correlation effects and disorder [2,3]. Here, we present results for the resistivity ρ of X-ray irradiated κ-$(ET)_2Cu[N(CN)_2]Cl$ in the temperature range from 5 K to 65 K and at various pressures up to 100 MPa. Measurements have been done on the same crystal for three different irradiation doses. The ρ(T) curves allow us to determine the metal to Mott insulator phase transition line TMI(p) and its evolution upon increasing degree of disorder. We find, that TMI(p) is shifted with increasing irradiation dose to lower pressures and that the critical end point of this line is reduced in temperature. Moreover, the jump in ρ(T) at the phase transition becomes reduced with increasing irradiation dose and eventually vanishes completely at a high level of disorder. The results will be discussed in the framework of disorder in Mott insulating materials.

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PI-53

Synthesis and electrical properties of new tetrathiafulvalene derivatives having chiral and hydrogen-bonding parts in the side chains

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Tetrathiafulvalenes (TTF) derivatives have been investigated as molecular metals in crystalline and supramolecular structures. One-dimensional stacks of CT complexes composed of TTF derivatives and acceptor molecules show highly electronic conductivities. Control of self-assembling processes can be formed the properties of molecular materials and devices built-up from functional small molecules. The assemblies of π-conjugated molecules have attracted special interest because of their great potential for the directional transportation of electrons and energy along with their molecular stacks. Molecular nanostructures are constructed intramolecular interactions such as CT or hydrogen-bonding interactions. Supramolecular approaches have a structural flexibility to form nanostructures of variety shape. Furthermore, helical fibrous assemblies have been observed by the introduction of chirality in molecule in the side chains. Chirality in molecules are key parts of self-assembling direction within the organizations. We previously demonstrated the helical structures of amphiphilic copper phthalocyanines. In this study, we report the electronic properties of several TTF derivatives and organic acceptors. We synthesized TTF derivatives having chiral and hydrogen-bonding side chains (Fig. 1). The synthesized molecules organized into helical and one-dimensional structures by using several intermolecular interactions. The films composed of (TTF derivatives) (acceptors) were casted on Si, quartz, HOPG and mica substrates. The surface morphology was examined by AFM and SEM. The vibration spectra of CT complexes in the cast film have showed the partially / fully charge states. The room temperature electrical conductivity of the cast films showed the semiconducting behaviors. The electrical conductivities measured by using PCI-AFM and magnetic properties will be discussed.

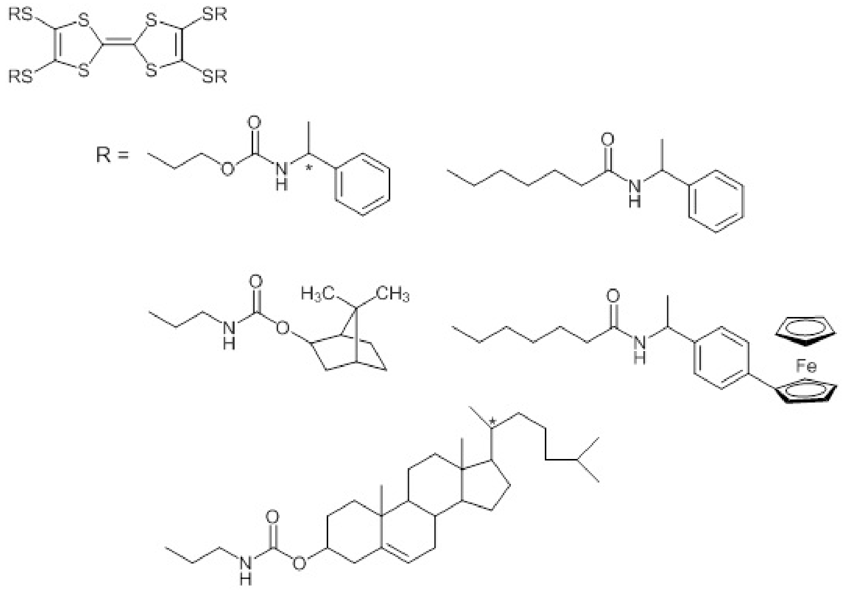

Figure 1: Chemical structures of several TTF derivatives.

Poster Session II

Thursday, September 10, 2015

20:00 - 22:00

PII-1

Dielectric anomalies in H_2O ice

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Through measurements of the complex dielectric constant of especially prepared samples of solid H_2O down to 7 K, three new anomalies have been identified. At Tp=100 K, a maximum was observed where proton (dis)ordering occurs. The broadened peak is attributed to the quenching of the disordered protons. At $T_{Ih\text{-}XI}$=60 K, a change in slope anomaly was identified which correlates to the structural phase transition from ice Ih (hexagonal) to ice XI (orthorhombic). At T_q=20 K, a minimum in the imaginary part of the dielectric constant was observed of which we attribute it to the onset of phase coherence of the ordered protons. This classifies H2O as another system that may exhibit macroscopic quantum phenomena. By repeating the same experiments up to hydrostatic pressures of up to 80 MPa as well as with D_2O, the phase stability region of ice XI in the P-T phase space is mapped out and the ice Ih/II/XI triple point is identified.

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PII-2

Ferroelectricity in fluorescent alkylamide-substituted pyrene derivative

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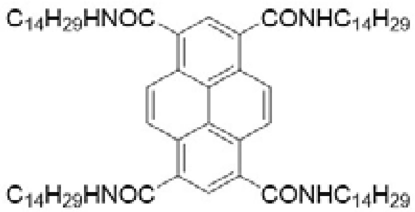
[2]*Tohoku univ., IMRAM, Sendai, Japan*

Intermolecular hydrogen-bonding molecular assembly, liquid crystalline (LC) phase, and ferroelectric properties of 1, 3, 5-trialkylamide-substituted benzene derivatives have been reported. On the other contrary, pyrene derivatives bearing long alkylester chains showed the LC phase. Herein, we successfully obtained a new pyrene derivative of N, N′, N″, N‴-tetra(tetradecyl)-1, 3, 6, 8-pyrenetetracarboxyamide (1), whose molecular assemblies, LC properties, dielectric responses, carrier transport properties, and optical properties were examined. Concentration-dependent fluorescence spectra in the chloroform showed the molecular aggregation properties of 1. The monomer emission with a maximum emission wavelength of ~400 nm was observed at dilute solution. Increasing in the concentration suppressed the fluorescence intensity around 400 nm and appeared a new emission band around ~500 nm at the concentration over 5×10^{-6} M. The latter band was assigned to the p-oligomer emission band. Since the excimer band of pyrene was observed at the concentration over 5×10^{-3} M, high molecular aggregation ability of 1 was confirmed through the p-stacking and hydrogen-bonding interactions. Molecule 1 also showed the hexagonal columnar LC phase, where the p-stacking and intermolecular N-H~O= hydrogen-bonding interaction of $C_{14}H_{29}$NHCO- chains played an important role to form the discotic LC phase. The polarization - electric filed (P-E) hysteresis curve of 1 was measured for the LC cell at 393 K (f = 0.5 Hz and E_{max} = 3.8 V/mm), which resulted in the ferroelectric behavior with coercive electric field of 2.6 V/mm and remnant polarization of 0.75 μC/cm^2. These dielectric switching phenomena were also associated with the dipole inversion of hydrogen-bonding chains along the p-stacking column.

Acknowledgement. Dr. S. Yamamoto and Prof. K. Mitsuishi at Tohoku University for the fluorescence lifetime measurement and Mr. Y. Wada and Prof. T. Takenobu at Waseda University for the I-V measurements.



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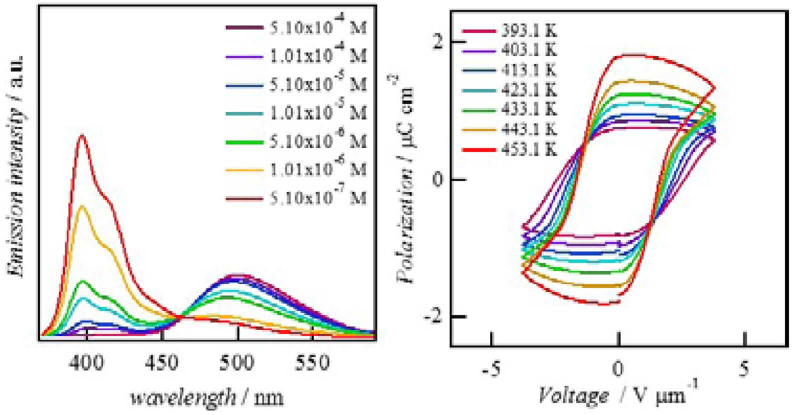

Figure: Chemical Structure of 1 (top), Concentration-Dependent Fluorescence Spectra of 1 (left), and P-E Hysteresis Curve of 1 (right)

PII-3

Pyroelectricity in an electronic ferroelectric crystal α-(BEDT-TTF)$_2$I$_3$ detected by temperature-modulation method

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An organic conductor α-(BEDT-TTF)$_2$I$_3$ shows a metal-to-insulator transition at 135 K (T_{CO}) due to charge ordering. We have demonstrated that the electronic transition is a ferroelectric one by an optical measurement, and focus on the physical properties for the interest into electronic ferroelectrics, a new class of ferroelectric material [1]. Ferroelectrics are characterized by significant external-field responses. According to a dielectric permittivity study [2], α-(BEDT-TTF)$_2$I$_3$ shows a sizable dielectric constant that remains down to far below T_{CO}. However, experimentally observed dielectric constant includes the contribution of free charge, preventing clear understanding of the role of the spontaneous polarization. To clarify the relationship between the polarization and the charge ordering, we measured pyroelectricity of the compound by adopting an ac method, modulating the sample temperature via periodical light irradiation. Figure 1 shows the electric current induced by the temperature modulation along the conductive plane. Note that the data observed at 0.5 Hz shows a plateau-form signal for $T>T_{CO}$, then a significant decrease of the signal around the transition point as lowering temperature. Though the observed electric current is dominated by the large plateau, the signal appearing in the metallic phase seems irrelevant to the ferroelectric polarization. Meanwhile, there exists a small anomaly denoted by the asterisk in Fig. 1. The nearby T_{CO} signal becomes prominent with lowering the temperature modulation frequency. We proved that the signal is a pyroelectric current associated with the spontaneous polarization due to the charge ordering by the analysis of the frequency dependence.

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[2] T. Ivek et al., PRL 104 (2010) 206406.

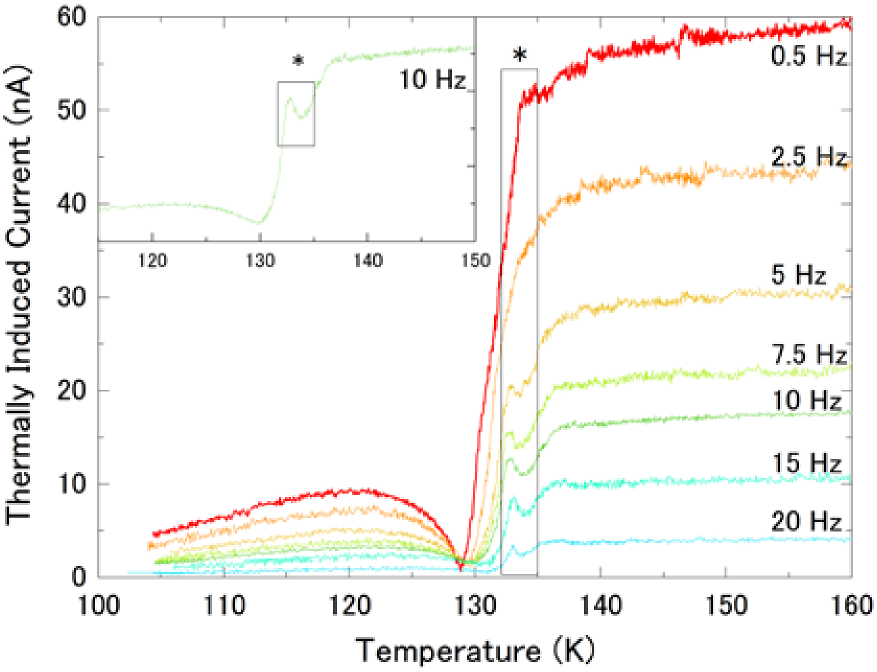

Figure 1: Thermally induced electric current in α-(BEDT-TTF)$_2$I$_3$ measured at various temperature-modulation frequencies.

PII-4

Dielectric properties of organic charge-transfer salts

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The EDT-TTF-based charge-transfer salts have attracted considerable attention in recent years due to their in many cases intriguing dielectric properties. One such example is κ-$(BEDT\text{-}TTF)_2Cu[N(CN)_2]Cl$. It was recently found to exhibit multiferroicity, for which a new electric-dipole driven mechanism of multiferroicity was proposed [1]. The polar moment in this system appears to be of predominantly electronic nature, arising from the dimerization of the BEDT-TTF molecules, combined with charge order. Another recent example showing interesting dielectric behavior is α-$(BEDT\text{-}TTF)_2I_3$, which shows relaxor-ferroelectric behavior deep in its charge-ordered state [2]. Here, we will present an overview of the dielectric properties of the above systems and provide new results on δ-$(EDT\text{-}TTF\text{-}CONMe_2)_2Br$ [3] and δ-$(EDT\text{-}TTF\text{-}CONMe_2)AsF_6$ [4]. At room temperature these compounds already exhibit charge order, but lack any dimerization. The Br-compound is a better conductor by more than two orders of magnitude at ambient conditions [5]. We find no indications of polar order in these systems, however there is evidence for two relaxational processes. In addition, large differences in the dielectric spectra along the three crystal axes were observed. The structural phase transition from orthorhombic to monoclinic seems to have no effect on the dielectric properties.

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Stochastic transitions between discrete quantum states of spin-soliton lattice in $[Mn((R/S)\text{-}pn)]_2[Mn((R/S)\text{-}pn)_2(H_2O)][Cr(CN)_6]_2$ chiral molecular magnet

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Series of the chaotic magnetization jumps good reproducible at the threshold magnetic field were observed in $[Mn((R/S)\text{-}pn)]_2[Mn((R/S)\text{-}pn)_2(H_2O)][Cr(CN)_6]_2$ single crystals during their demagnetization. Figure 1 shows fragments of the demagnetization curves M(H) recorded at the same conditions at the temperature 8 K. Critical magnetic field values corresponding to start H_{C1} and finish H_{C2} of the magnetization jump series are shown by arrows. Theoretical demagnetization curves [1] between which magnetization jumps occur are shown by dashed lines. The correlation of temperature dependencies of the magnetic field threshold value (H_C) and magnetic anisotropy field indicates contribution of the magnetic anisotropy in non-linear spin-soliton structure. Competition of the Dzyaloshinskii-Moria asymmetric exchange interaction and magnetic anisotropy provides new kind of spin excitations which stability depends on external magnetic field [1].

The work was supported by the RFBR grant 15-02-05149.

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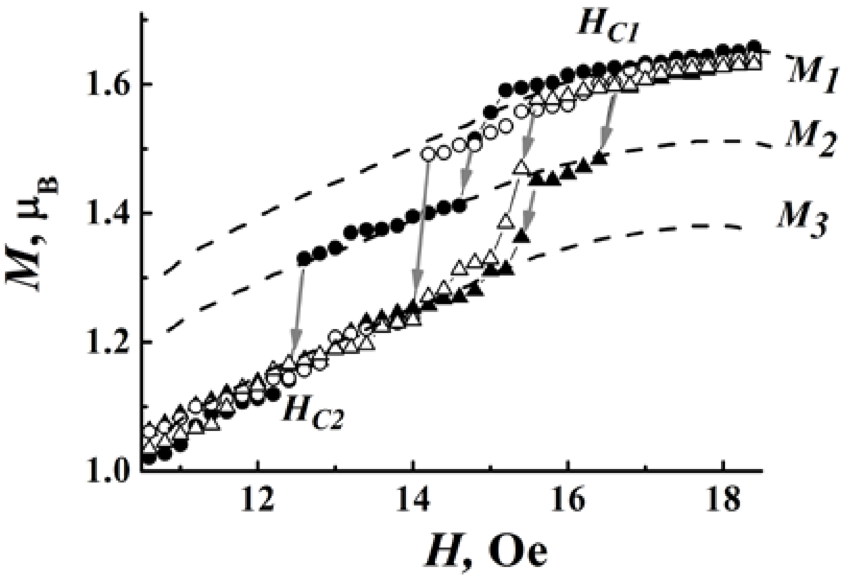

Figure 1: Fragments of the demagnetization curves M(H)

PII-6

Low-energy excitations in the quantum spin liquid κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$

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The suppression of long range magnetic order due to geometrical frustration gives rise to the quantum spin liquid state. Theoretical considerations predict enhanced absorption within the Mott gap caused by spinons, which results in a low-frequency power-law behaviour of the optical conductivity, i.e. for $\hbar\omega_c < J \approx 250$ K [1]. To verify this hypothesis, the optical conductivity of the spin liquid candidate κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$ was measured, where the dimerized organic molecules are arranged on a triangular lattice. An extremely wide energy range from radio frequencies up to the near infrared (10^{-13} eV – 1 eV) was covered by dielectric spectroscopy, THz absorption and optical reflectivity measurements. We could indeed identify a power-law behaviour $\sigma(\omega) \propto \omega^\beta$ where two distinct exponents β change from 0.9 to 1.4 at low temperatures, with the corresponding crossover scaling with temperature: $\hbar\omega_c \approx kBT$ [4]. Similarly, a power-law exponent of 1.4 was observed for the Herbertsmithite $ZnCu_3(OH)_6Cl_2$, another two-dimensional spin liquid candidate with Kagome lattice structure [2]. Comparing these values to the theoretically predicted exponents of 2 and 3.3 reveals significant discrepancy [1]. Hence, these experimental findings may motivate a refinement of the theoretical framework.

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[3] S. Elsässer et al., Phys. Rev. B 86 (2012) 155150 [4] A. Pustogow et al., http://arxiv.org/abs/1412.4581

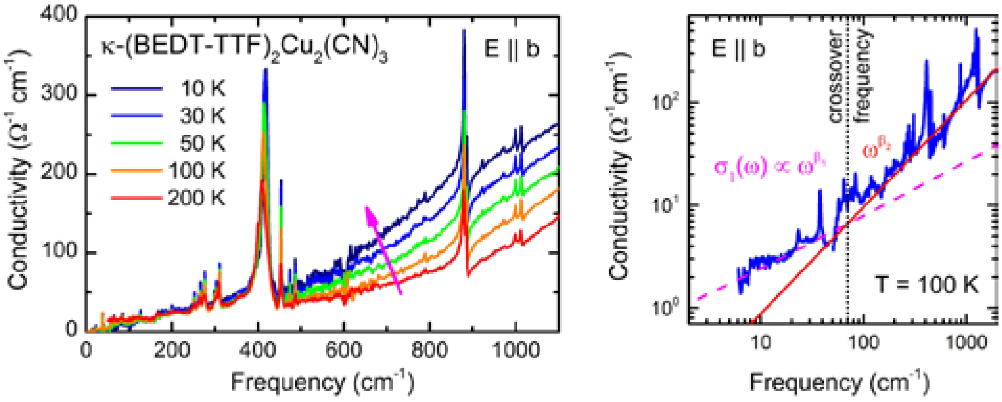

Figure: (a) Unlike a typical insulator the quantum spin liquid compound exhibits increasing absorption towards low temperatures, as already pointed out in [3]. (b) The optical conductivity follows two distinct power laws with exponents β1 and β2.

Magnetism in a family of S = 1 square lattice antiferromagnets $NiX_2(pyz)_2$ (X = Cl, Br, I, NCS; pyz = pyrazine)

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We describe four Ni(II) coordination polymers based on Ni--pyz--Ni square-lattice layers: $NiX_2(pyz)_2$ (X = Cl, Br, I, NCS; pyz = pyrazine). The results of measurements of synchrotron x-ray powder diffraction, electron-spin resonance, muon-spin relaxation, heat capacity and magnetization are presented. Together these experimental results paint a picture of a family of structurally similar materials whose magnetism is altered significantly by the variation in axial ligand. The size and anisotropy of the antiferromagnetic exchange interactions, the presence of zero-field splitting and long-range magnetic order are all affected by the choice of X. We discuss these findings in comparison to other Ni(II) square-lattice materials and low-dimensional magnets with halogen-containing ligands.

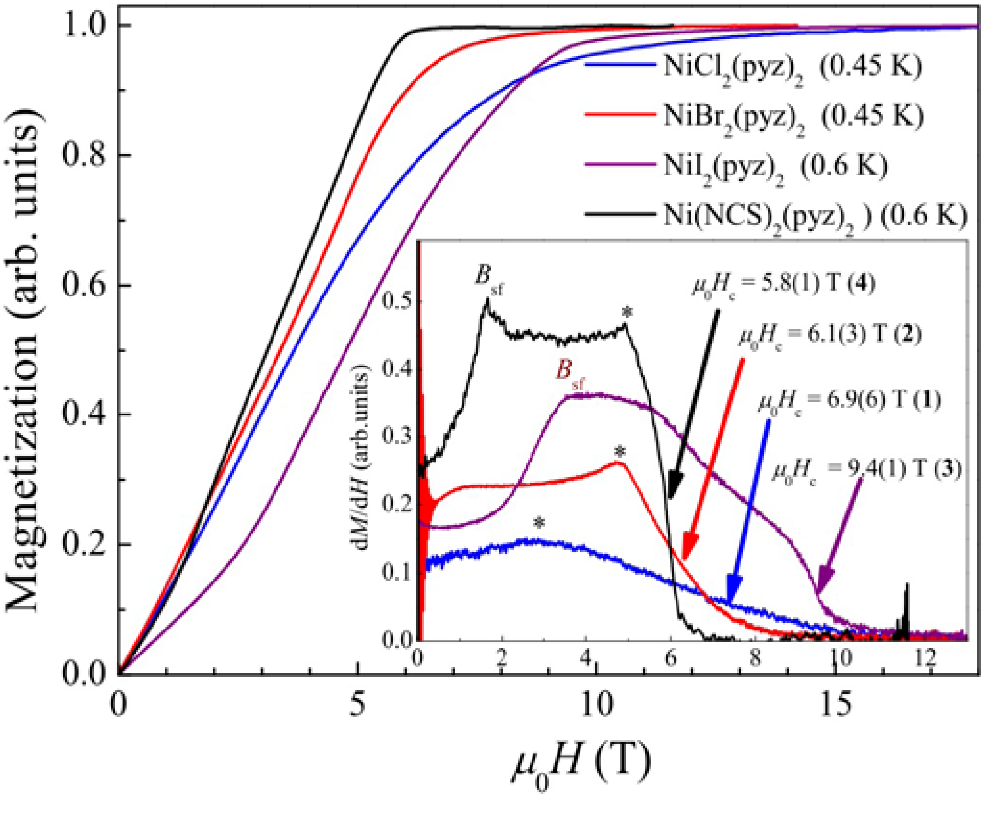

Figure 1: Low-temperature, pulsed-field magnetization data. Inset: field derivative of magnetization showing saturation fields

PII-8

The magnetic ground state of two isostructural polymeric quantum magnets investigated with neutron diffraction

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Low-dimensional magnetic materials exhibit phenomena which cannot be explained semi-classically. One example is quantum fluctuations of magnetic moments at low temperatures, necessitated by the Heisenberg uncertainty principle [1]. For a square lattice S = 1/2 Heisenberg antiferromangnet, quantum-Monte-Carlo simulations predict that quantum fluctuations reduce the ordered moment per ion from its classical value [2]. Coordination polymers, in which the magnetic exchange between metal ions is mediated by organic molecules, offer the means to test the properties of low-dimensional magnets by constructing 3D lattices of ions with anisotropic exchange pathways. We present neutron powder diffraction experiments which compares the magnetic ground state of two coordination polymers: (i) $[Cu(HF_2)(pyz)_2]SbF_6$ (where pyz = pyrazine), a quasi-2D magnet based on planes of S = 1/2 moments arranged on a square lattice [3], and (ii) a new, isostructural polymer $[Co(HF_2)(pyz)_2]SbF_6$. We find the ordered moments of the Heisenberg S = 1/2 Cu(II) ions in $[Cu(HF_2)(pyrazine)_2]SbF_6$ are 0.60 ± 0.10 μB, whilst the ordered moments for the Co(II) ions in [Co(HF2)(pyrazine)2]SbF6 are 3.17 ± 0.09 μB. For Cu(II), this reduced moment indicates the presence of quantum fluctuations below the ordering temperature. We show from heat capacity and ESR measurements, that due to the crystal electric field splitting of the S = 3/2 Co(II) ions in $[Co(HF_2)(pyrazine)_2]SbF_6$, this isostructual polymer also behaves as an effective spin-half magnet at low temperatures. The Co material shows evidence for significantly more isotropic magnetic exchange, strong easy-axis anisotropy and neutron diffraction data which do not support the presence of quantum fluctuations in the ground state.

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PII-9

μSR study of quasi two-dimensional S = 1/2 triangular antiferromagnet, EtMe$_3$Sb[Pd(dmit)$_2$]$_2$

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Magnetic ground states of quasi-two-dimensional (Q2D) triangular Heisenberg antiferromagnetic (AF) systems are of great interest. Magnetic frustration arising from the triangular exchange network suppresses the AF order. This kind of quantum-spin states without either long-range magnetic order or lattice symmetry breaking is named quantum spin liquid (QSL) state. Although experimentalist have sought real model materials with QSL state for quite some time, only a few candidate materials are known to this date. Recently, we have performed longitudinal field (LF) μSR measurements on a QSL candidate, EtMe3Sb[Pd(dmit)$_2$]$_2$ which do not show any AF order due to strong spin frustrations[1]. Preliminary analysis suggest that magnetic field H_{ext} dependence of muon relaxation rate, λ is proportional to $H_{ext}^{-1/2}$ behavior in a field range of $0.1 \leq \mu_0 H_{ext} \leq 100$ mT at low-temperatures. Such a behavior is expected from spins diffusing along a one-dimensional direction (see figure).

[1] T. Itou et al., Phys. Rev. B, 77, 104413 (2008)

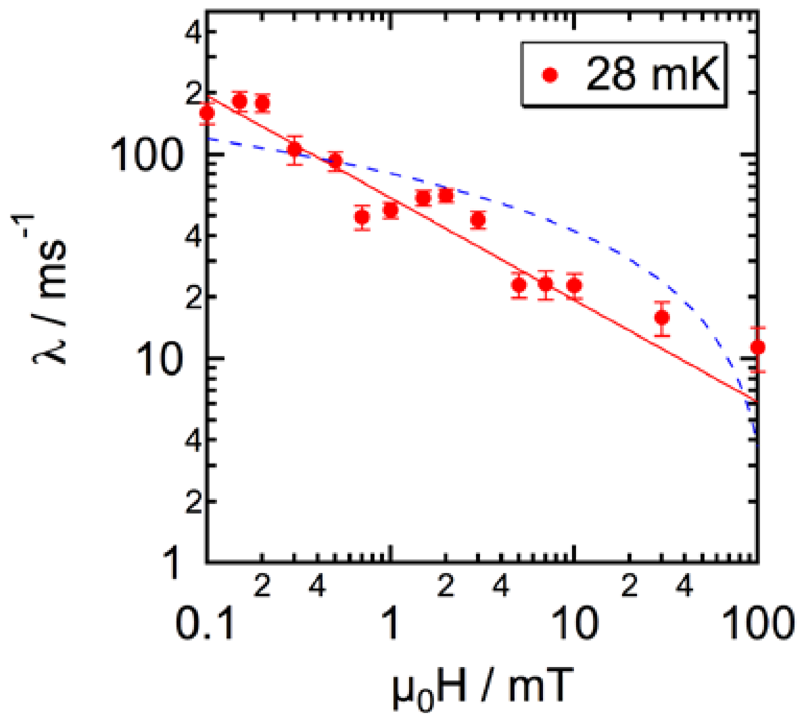

Figure 1: Magnetic-field-dependent λ of EtMe3Sb[Pd(dmit)2]2 measured at 28 mK. The lines are best-fit curves for 1D(solid) and 2D(dashed) models.

PII-10

Magnetic properties of organic radicals based on carboxyphenyl-, furyl- and thienyl-substituted nitrony nitroxide and iminonitroxide

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Magnetic properties of organic radicals based on carboxyphenyl-, furyl- and thienyl- substituted nitronyl nitroxide (NN) and iminonitroxide (IN) were investigated by measuring temperature and magnetic-field dependences of magnetization. The magnetic behavior of the radical 4-carboxy-2-hydroxyphenyl nitronyl nitroxide (4-CA-2-HPNN) is interpreted in terms of the one-dimensional (1-d) ferromagnetic (FM) Heisenberg model with the coupling constant $J/k = +0.40$ K above 10 K. Below 10 K, however, antiferromagnetic (AFM) interaction with $zJ'/k = -0.42$ K between neighboring 1-d chains reduces magnetization to some extent. Many other carboxyphenyl NN studied here exhibit magnetic behaviors interpreted in terms of the 1-d alternating AFM Heisenberg model. The magnetic behavior of the radical 2-benzo[b]thienyl nitronyl nitroxide (2-BTHNN) is interpreted in terms of the two magnetic sublattices model in which one sublattice shows 1-d intermolecular FM interaction with $J/k = +0.06$ K and the other sublattice exhibits the spin-dimer intermolecular AFM interaction with $J'/k = -55$ K. The finding of the coexistence of FM and AFM magnetic sublattices is consistent with the crystal structure of 2-BTHNN. To discuss magnetism in 4-CA-2-HPNN and 2-BTHNN, magnetic behaviors of other NN and IN derivatives are shown and compared to each other.

PII-11

Anomalous magnetic properties around the QSL phase in Pd(dmit)$_2$ salts

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A frustrated quantum spin system with a two-dimensional (2D) triangular lattice is formed in a series of anion radical salts (Cation+)[Pd(dmit)2]2, where dmit denotes 1,3-dithiol-2-thione-4,5-dithiolate. In the crystal, spin-1/2 units, [Pd(dmit)$_2$]$_2$-, form 2D isosceles triangular structure, which is characterized by a frustration parameter t′/t; t and t′ are interdimer transfer integrals. Especially, EtMe$_3$Sb[Pd(dmit)$_2$]$_2$, t′/t of which is nearly 1, has attracted considerable attention, because it has no magnetic order down to 19 mK (< 0.01% of antiferromagnetic exchange interaction), namely a quantum spin liquid (QSL) state is formed. The frustration parameter can be tuned by changing counter cation, and the ground state varies with t′/t. The QSL is situated between an antiferromagnetic long range order phase (AFLO) and a charge ordering phase (CO). Recently, we succeeded in fine control of t′/t by using mixed cation salts, which will lead us to reveal boundary behavior around QSL. In this study, we measured magnetic susceptibility of mixed cation salts with finely controlled t′/t and found several anomalous magnetic properties around the QSL state. Transition temperatures (TN and TCO) of the pristine salts smoothly decreased with changing t′/t and QSL was observed in a specific range of t′/t, namely, QSL appeared as a phase. Some magnetic spins, which obeyed Currie's law, were always observed and the number of them showed systematic change depending on t′/t. The number of such "free spins" is constant in the QSL phase and about 10 times larger than that in the AFLO phase. We also observed anomalous magnetic anisotropy in the AFLO state near the QSL phase.

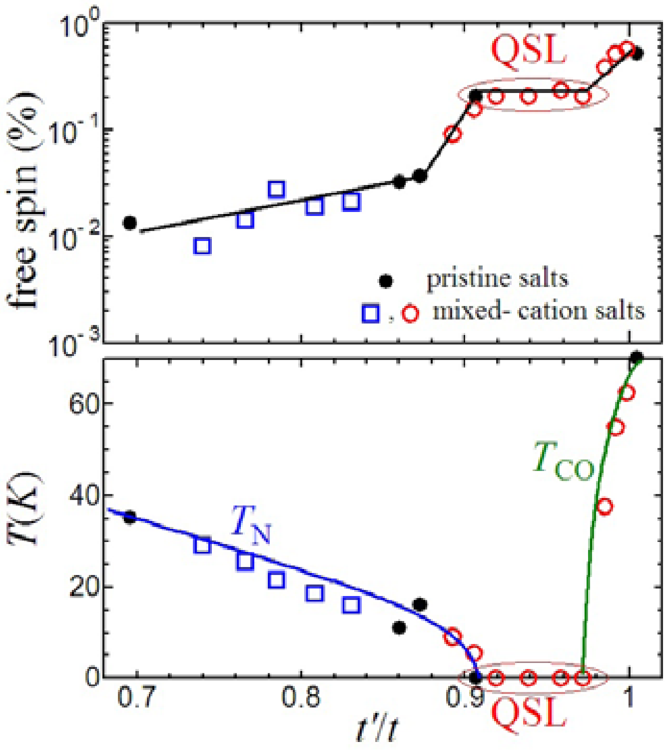

Figure: Phase diagram (lower panel) and free spin concentration (upper panel) of (Cation)[Pd(dmit)$_2$]$_2$

PII-12

Frustration effect on the antiferromagnetic ordering in quasi triangular localized spin systems, β′-$Et_2Me_2P[Pd(dmit)_2]_2$ and β′-$Me_4P[Pd(dmit)_2]_2$

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β′-$X[Pd(dmit)_2]_2$ ($X=Me_4P^+$, $Et_2Me_2P^+$, etc and dmit=1,3-dithiol-2-thione-4,5-dithiolate) are known as two dimensional (2D) Mott insulators in which a spin-1/2 localizes on each $Pd(dmit)_2$ dimer. These salts are considered to be a frustrated spin system due to quasi-triangular dimer arrangement in the 2D layer. The close correlation between anisotropy of the triangle and antiferromagnetic transition temperature TN is the characteristic point of this system. The larger anisotropy of the triangle in the Me_4P salt (TN=42K) is considered to reduce the frustration leading to the higher ordering temperature than in the Et_2Me_2P salt (TN=17K). In order to clarify the magnetic structures of these salts and to figure out the effect of frustration systematically, we measured and analyzed the angular dependence of ^{13}C NMR spectra in the AF phase. The external field of 7.0~8.3T, much larger than the spin-flop field of ~1.0T, was rotated in the crystallographic ac*- and bc*-plane. The angular dependence of observed peak positions were compared with the calculated ones with the use of hyperfine coupling constant, A_{aniso}~293Oe/μB, which are derived experimentally. Quite similar results are obtained by using the electron distributions given by the extended Hückel MO calculation. We found that the results for both salts are very well explained by assuming that the easy and the hard axes are the c* and the b-axis, respectively. The localized moment were found as small as 0.28 μB/dimer for the Et_2Me_2P salt, while 0.45 μB/dimer for the Me_4P salt. We believe that the extremely small value of the localized moments in the Et_2Me_2P salt should reflect the strong frustrations.

^{13}C-NMR investigation on the spin state of the neutral-ionic transition material TTF-CA

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The organic charge transfer complex, TTF-CA, is well known to undergo the neutral-ionic (NI) phase transition, which is characterized by charge transfer and lattice dimerization. In many cases, the charge transfer occurs simultaneously with lattice dimerization so that NI transition materials are non-magnetic insulators in both neutral (closed shell state) and ionic (spin-Peierls state) phases. In TTF-CA, however, the recent 35Cl-NQR investigations have indicated that a non-dimeric ionic phase emerges at high pressures above 9 kbar [1], suggesting a paramagnetic phase. It is puzzling that nevertheless the activation of the ag mode, indicative of the dimerization in the ionic phase, was observed under pressures by infrared absorption measurement [2]. The spin and lattice states in TTF-CA at high pressures are intriguing. In the present work, we have investigated the spin state in TTF-CA under pressures by 13C-NMR. At room temperature, as pressure is applied, the spectral shift (= Knight shift + Chemical shift) and the spin-lattice relaxation rate, 1/T1, increase, providing microscopic evidence for the spin degrees of freedom becoming vital; at 14 kbar (H || a), for example, the Knight shift, which is proportional to the magnetic susceptibility, increases up to 25 ppm (Fig. 1). However, the value of shift is too small to be explainable in terms of the 1D uniform Heisenberg model with the reported band parameters and 1/T1 is unexplainable as well. These NMR features suggest an alternative picture of the paramagnetic state; that is, solitonic and mobile spin excitations in an ionic dimer liquid, which also reconciles with the optical results.

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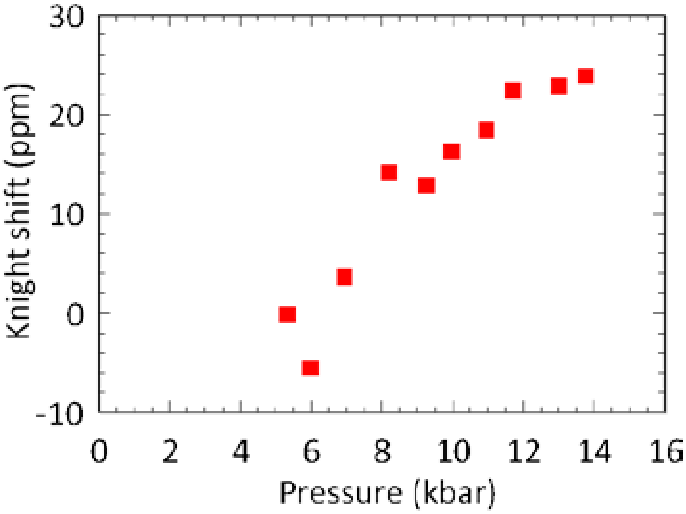

Figure 1:
Pressure dependence of the Knight shift in TTF-CA

PII-14

Optical study of an anisotropic triangular lattice material, κ-$(BEDT\text{-}TTF)_2B(CN)_4$

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We studied the electronic states and their anisotropy in triangular spin-lattice BEDT-TTF compounds using optical methods. The triangular nature and frustration effects in such materials have been discussed mainly based on the ratio of two transfer integrals, t and t′. They are generally determined not by electronic measurements but by the combination of the structural data and calculated methods. In this study, we investigated the optical properties of κ-$(BEDT\text{-}TTF)_2B(CN)_4$, which has an anisotropic triangular lattice and whose t′/t is calculated to be beyond 1.4 from the structural data at room temperature [1]. The infrared charge transfer transitions of κ-$(BEDT\text{-}TTF)_2B(CN)_4$ show a larger anisotropy in comparison with those of the other κ-type BEDT-TTF compounds. This suggests that the observed anisotropy reflects the electronic anisotropy. Moreover, the optical anisotropy in the infrared region shows a clear temperature dependence, which seems consistent with the temperature dependence of t′/t calculated from the structural data. The electronic states of κ-$(BEDT\text{-}TTF)_2B(CN)_4$ in the low temperature region are discussed from the infrared and Raman scattering spectra.

[1] Y. Yoshida et al., submitted.

PII-15

Magnetic field induced fluctuations in a quantum spin liquid κ-$(BEDT\text{-}TTF)_2$-$Ag_2(CN)_3$ seen by high-frequency electron spin resonance spectroscopy

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Magnetic ordering in strongly interacting systems might be washed out by strong quantum fluctuations. The almost isotropic two-dimensional triangular magnetic lattice of spin-1/2 dimers makes the κ-$(BEDT\text{-}TTF)_{2\text{-}}X_2(CN)_3$ material's family (X=Cu, Ag) a prime example for the long-sought quantum spin liquid state (QSL) [1]. The presence of frustration leads to macroscopic ground-state degeneracy, preventing that long-range order set in. The destruction of magnetic order is further aided by low-dimensionality and low spin values. The absence of magnetic ordering has been proven in κ-$(BEDT\text{-}TTF)_{2\text{-}}Cu_2(CN)_3$ down to 20 mK [2]. Recent muon spin rotation experiments [3] in agreement with numerical models [4] have shown, however, that the QSL state at low temperatures and high magnetic fields is sensitive to small spin-space anisotropies, such as the anisotropy due to the Dzyaloshinskii-Moriya (DM) interaction. Here we investigate κ-$(BEDT\text{-}TTF)_{2\text{-}}Ag_2(CN)_3$ using multi-frequency electron spin resonance (ESR) spectroscopy. We follow the ESR line of the S=1/2 ET_2 dimers in magnetic fields up to 15 T and down to 1.5 K in temperature. In directions non-parallel with the DM vector, we observe a quadratic in field broadening and shift of the ESR line upon applying the magnetic field. We propose that the field-induced static and fluctuating magnetizations arise from the interplay of the DM and Zeeman interactions. The study of these magnetic fluctuations gives insight into the relevance of the DM interaction in the high-field induced quantum critical phase of the QSL state.

[1] L. Balents, Nature 464, 199-208 (2010)
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PII-16

New structural features in the spin-liquid system κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

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One of the most intriguing state observed in strongly correlated systems is the spin-liquid state, characterized by the absence of magnetic ordering in spite of a strong antiferromagnetic coupling. Most prominent spin-liquid system is the κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ salt, built with a triangular network of BEDT-TTF (ET) dimers, separated by planar polymeric network of Cu ions and (CN) groups. The symmetry of the structure is non polar with a disorder inside the polymeric group. Due to the triangular geometry, one expects a magnetic frustration. Indeed, even if the exchange coupling between molecules is of about 250 K, there is no magnetic order reported down to 30 mK [1]. Although there are no possible electric dipoles on the ET molecules due to the non polar symmetry of the structure [2], the fingerprints of relaxor ferroelectricity have been established by dielectric spectroscopy [3]. This result demonstrates an heterogeneity which has been related to domains whose formation was associated with the CN groups disorder. To understand these findings in relation with the spin-liquid phase, it is important to reinvestigate the structural properties of the system. We have recently performed an X-ray scattering study and have demonstrated that the 300K structure is not the one claimed until now. Indeed, we have detected the presence of Bragg reflections forbidden in the conventional space group. The presence of these reflections rules out the structure proposed until now. Our analysis shows that the room temperature structure can be polar with a partial ordering of the CN groups. The study of the evolution of the structure with temperature is in progress and should strongly help to understand the possible ferroelectric-like behaviour and to shed light on the complex spin-liquid state.

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Formation of π-π stacking of cationic coronene

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Highly symmetric (D_{6h}) polyaromatic hydrocarbon, coronene, has attracted a great deal of attention recently because of its relevance to graphene. A recent outstanding result is the discovery of a superconducting phase below 15 K in potassium-doped coronene [1]. Although its crystal structure remains unknown, it must involve segregated columns or layers of anionic coronene to produce the π-conduction pathway. Such an assembly via π-π interactions has attracted much interest not only from the perspective of materials science but also in the fields of astrophysics and physiology. Very recently, we obtained the first coronene cation radical salts formed with dianionic molybdenum halide clusters, $(\text{coronene})_3\text{Mo}_6\text{X}_{14}$ (X = Cl and Br) [2]; however, the salts show semiconducting behavior possibly because of the lack of π-π stacking interactions as well as the presence of charge-disproportionation. In this study, we succeed in obtaining the first coronene cation radical salts with columnar structure, by electrocrystallization in the presence of Lindqvist-type polyoxometalate dianions [3]. In the $(\text{coronene})_3\text{M}_6\text{O}_{19}$ (M = Mo and W) salts, there are significant π-π interactions within the coronene column, which suppress the in-plane molecular rotation of coronene molecules and allow structural observation of a static Jahn-Teller distortion associated with splitting of the doubly degenerate e_{2u} HOMO. The fairly homogeneous charge state (ca. +2/3) of all coronene molecules in the present salts was confirmed by Raman spectroscopy and tight-binding model analysis. Although non-uniform stacking leads to band insulating behavior because of the energy gap opening at the Fermi level, the present salts include an unprecedented columnar structure of partially charged coronene molecules.

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PII-19

Carrier doping into a Mott insulator κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl using electric double layer transistor

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Carrier doping into certain types of Mott insulator can cause an insulator-to-metal transition and superconductivity. The phase diagrams for the electron- and hole-doped Mott insulators differ in detail and the doping asymmetry has been of interest. To study the asymmetry, we should prepare a non-doped Mott insulator and inject electrons or holes to compare each doping effect. However, most of the doped Mott insulators such as high-T_c superconductors are either only hole- or electron-doped, and the parent materials are usually different. k-type BEDT-TTF salts are suitable for examining this issue because they are originally non-doped and a method of field-effect doping is applicable. In this study, we applied the principle of the electric-double-layer transistor (EDLT) to a Mott insulating salt κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl (κ-Cl). The doping asymmetry on the resistivity and the Hall effect was investigated. The EDLT device was fabricated with laminating a thin single crystal of κ-Cl and a gate electrolyte on a polymer substrate. We observed a clear ambipolar field effect where even metallic conduction appeared under electron doping. At low temperature, the Hall coefficient upon both electron and hole doping indicated the presence of delocalized carriers, which is much more than the injected carriers. However, the Hall coefficient was several times larger on the hole-doped side than on the electron-doped side. One possible scenario is that the intrinsic carriers are partially delocalized under hole doping, while most of them are delocalized under electron doping (recovery of the Fermi surface).

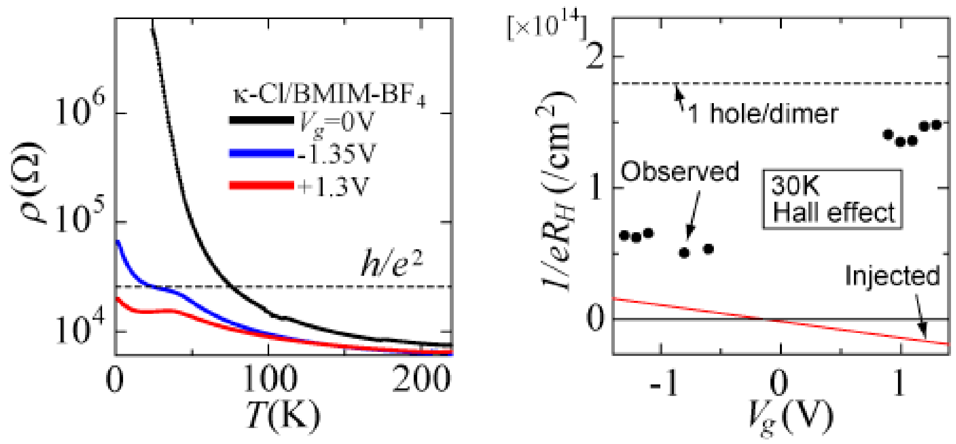

Figure: (Left) Temperature dependences of the sheet resistivity with and without gate voltage. (Right) Gate voltage dependence of 1/eRH at 30K. The red line indicates the density of the injected carriers estimated from the capacitance of the gate electrolyte.

PII-20

Formation and characterization of self-assembled monolayers on ferromagnetic surfaces for spintronic applications

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In the field of organic spintronics, the surface modification of ferromagnetic (FM) electrodes is a key point in order to study and improve their properties and performance.[1] In this scenario, one possibility is the formation of self-assembled monolayers (SAMs) on their surfaces. SAMs are thin films formed on a surface by the spontaneous adsorption of molecules from solution (or gas phase) giving rise to new inorganic-organic interfaces with new electronic and chemical properties.[2] Their main features are: defined structure, lightweight, flexible, nanometer thick and their properties can be tailored by chemical synthesis. This allowed the use of SAMs in spintronic devices, like spin tunnel junctions.[3] There are few reports about the growth of SAMs on FM surfaces and they are mainly developed on Cobalt and Nickel.[4] Apart from them, another relevant FM material, currently used in spintronics and magnetic storage technology, is Permalloy (Py), for whom no functionalization has been reported. This material is a nickel-iron alloy that shows interesting magnetic properties. When exposed to air, like other FM metals, Py develops a thin oxide layer, which passivates the surface and protects the bulk from the oxidation. In this work we will present for the first time a methodology to graft on Permalloy different types of SAMs with different functional groups. The formed monolayers have been fully characterized by means of atomic force microscopy, X-ray photoelectron spectroscopy, matrix assisted laser desorption ionization time-of-flight mass spectrometry, infrared reflection absorption spectroscopy. Moreover using microcontact printing, a soft lithography technique, we obtained well-defined SAM patterns on Py that could be used as a protecting layers when performing a wet chemical etching on permalloy, as shown in Figure 1.

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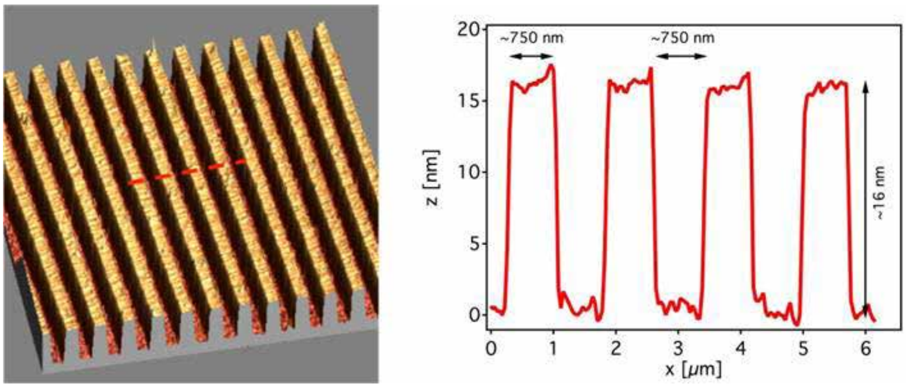

Figure 1:
AFM topographic image of some Py stripes formed on a solution etched ~16 nm thick Py substrate micropatterned with a C18P SAM: (left) 3D image of 20 µm x 20 µm and (right) height profile corresponding to the red line in (a).

PII-21

Structure analysis of cryolitic melts with a molecular dynamics simulation

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It has been shown how it is possible to simulate the Raman spectra of moderately complex molten salts,in previous works [1,2]. These works presents a way of bridging an interpretative gap between the structures as seen spectroscopically and that is understood from diffraction experiments. In spectroscopy, attention is focused on the coordination complexes responsible for discrete Raman bands seen in the spectra of melts with polyvalent cations[3], which are similar to those seen in a molecular system, however in diffraction experiments, the data is discussed from a more atomistic perspective in terms of radial correlations in an ionic mixture[4,5]. In this work, the structure analysis of cryolitic melts has been calculated from molecular dynamics computer simulations using a polarizable ionic potential obtained by force-fitting to ab inito electronic structure calculations. Simulations which made use of this ab initio derived polarizable interaction potential reproduced the structure and dynamical properties of crystalline cryolite. The transferability of the potential model from solid state to the molten state is tested by comparing results for the Raman spectra of melts of various compositions with those previously obtained with empirically developed potentials and with experimental data. The shapes of the spectra and their evolution with composition in the mixtures conform quite well to those seen experimentally, and we discuss the relationship between the bands seen in the spectra and the vibrational modes of the of the $AlF_n^{(3-n)}$ coordination complexes which are found in the NaF/AlF_3 mixtures. The simulations thus provide a link between the structre of the melt as derived through Raman spectroscopy and through diffraction experimens.It will be showed results for quantities which relate to the degree of cross-linking between these coordination complexes and diffusive properties of ions.

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PII-22

Charge disproportionation in two-band extended Hubbard model

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Motivated by recent reports of ferroelectric behavior in a few charge-transfer salts [1], we investigate possible charge disproportionation patterns in the framework of a simplified two-band extended Hubbard model at 3/4 filling. We shall present the Hartree-Fock as well as the variational Monte Carlo results for the phase diagram of this model as a function of the onsite Coulomb interaction U, the intra-orbital Coulomb interaction U‘, and the nearest-neighbor interaction V.

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PII-23

Coexistence of spin-density wave and superconductivity in $(TMTSF)_2ClO_4$ probed by nonlinear

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It is now well established [1-4] that in Bechgaard salts spin-density wave (SDW), near its critical point, coexists with small superconducting (SC) domains elongated, counterintuitively, in the direction of the weakest conductivity. The mechanism behind the formation of such unusual spatial texture is still being debated [1,3,4], with the two approaches, soliton phase theory [5] and macroscopic segregation [4], being most promising. However, neither of them consistently accounts for all the observed effects in the coexistence region [3]. In soliton phase the domains can be imagined as a set of “defects” of the SDW order parameter and hence should be affected by the collective motion of density wave, whereas in phase segregation scenario the domains should be intact. Following this idea we perform nonlinear conductivity measurements in $(TMTSF)_2ClO_4$ to probe the nature of SC/SDW coexistence. In the this region we observe new effect: drastic decrease of conductivity with increasing electric field; the effect demonstrating quite weak temperature and magnetic field dependence. Conductivity decrease presents exclusively in the coexistence region: it is most prominent in the vicinity of homogeneous SDW (h-SDW) and vanishes as SDW disappears. Furthermore, an admixture of phase segregation is evident from our data in the close vicinity of SDW endpoint and vanishes towards h-SDW, whereas SC/SDW coexistence persists. We discuss possible links of the observed effect with the realization of soliton phase in Bechgaard salts.

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New electroactive 2-(1H-Pyrrol-1-yl)pyrimidines containing chromophore moieties: synthesis, optical and electrochemical properties

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[3]*Natural Sciences Institute. Perm State University, Russia*

In search for materials with improved electronic properties useful for the purposes of organic electronics various compounds including in their conjugated systems such electron-deficient heterocycles as pyridine, pyrimidine, quinoline, quinoxaline, triazine are intensively synthesized. Incorporation of these heterocycles into the structure of future materials is frequently used to balance the charge injection from electrodes in light-emitting devices and solar cells. The same approach is often practised with an aim to tune luminescent properties of compounds in solutions and solid state as well. The insertion of pyrimidine moiety is noteworthy at least for two reasons: (i) this heterocycle has a high value of electron affinity [1] and (ii) due to the lack of ortho ortho interaction of C-H hydrogenes in the 2C-aryl substituted pyrimidines having wherefore more planar structures that seriously improves conjugation degree in the system [2,3]. We have synthesized a set of novel substituted 2-pyrrolopyrimidines of Ar(Ar'')AAr' general formula which possess an extended and branchy network of conjugation, where A is a pyrimidine core, Ar and Ar' some electron excessive substituents (combination of various carbo- and heterocycles such as thiophene, carbazole, phenothiazine, oxadiazole) and Ar'' stands for the pyrrole ring. Cyclic voltammetry was used to study their electrochemical properties as well as to estimate their capability to be electrochemically polymerized. UV/vis spectra and spectra of fluorescence were determined for all the newly synthesized compounds, the values of HOMO, LUMO energies and Egopt and Egel were experimentally calculated (Fig. 1).

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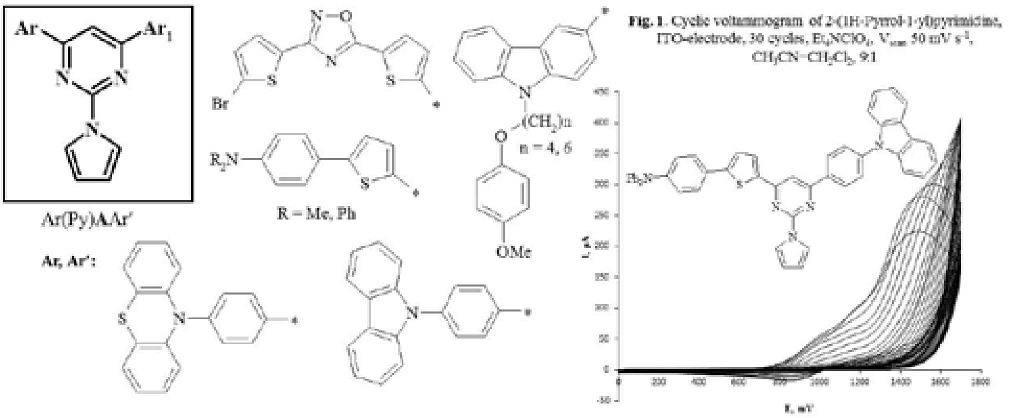

Figure 1:
Cyclic voltammogram of 2-(1H-Pyrrol-1-yl)pyrimidine, ITO-electrode, 30 cycles, Et_4NClO_4, Vscan 50 mV s-1, $CH_3CN–CH_2Cl_2$, 9:1

PII-25

Orbital-selective NEXAFS: origin of the charge transfer in single crystals based on thiophene derivatives

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Using orbital-selective NEXAFS (near edge X-ray absorption fine structure) analysis it was possible to elucidate the spectroscopic fingerprints of the formation of charge transfer (CT) phases in the novel organic compounds DTBDT-TCNQ and DTBDT-F_4TCNQ, based on dithieno[2,3-d;2',3'-d']benzo[1,2-b;4,5-b']dithiophene donor. Single crystals of high quality were obtained by physical vapor transport and characterized by X-ray diffraction and IR-spectroscopy. Direct comparison of nitrogen and fluorine K-edge NEXAFS spectra of the compounds and the pure donor and acceptor moieties provide precise information on specific orbital occupations and their change. Taking into account theoretical calculations on possible transitions when forming the CT complex, we show that tuning of the acceptor strength from TCNQ to F_4TCNQ dramatically changes the mechanism of charge transfer and the participating orbitals. The fingerprints in the NEXAFS spectra are very pronounced allowing for a deep look into the richness of the CT mechanism.

The project is funded through Transregio SFB TR 49 (Frankfurt, Mainz and Kaiserslautern), Graduate School of Excellence Materials Science in Mainz MAINZ and by Centre for Complex Materials COMATT, Mainz.

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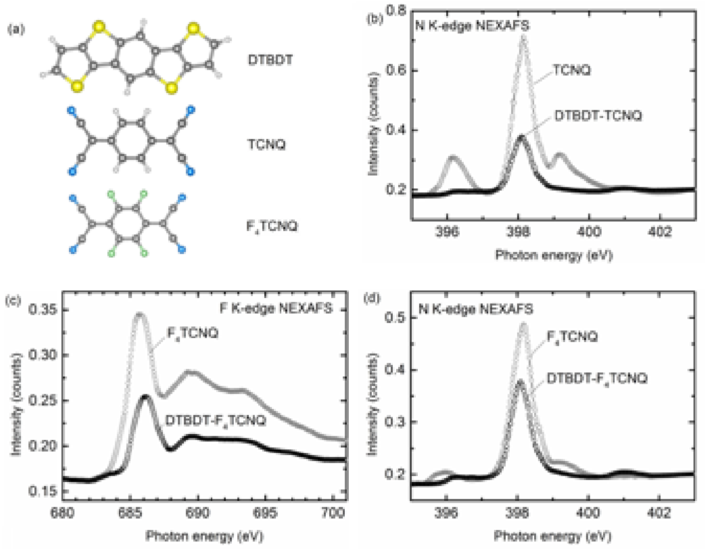

Figure 1: Molecular structure of donor DTBDT and acceptors TCNQ and F4TCNQ (a), N K-edge spectra of DTBDT-TCNQ (b), DTBDT-F_4TCNQ (c) and F K-edge NEXAFS of DTBDT-F_4TCNQ (d). All NEXAFS spectra are shown in comparison to pure acceptor on same intensity scale.

New structural units for magnetic and conducting materials based on $[M(pztdt)_2]$, M=Ni, Pd, Pt

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Aiming at enlarging the our family of more extended bisdithiolene [1,2] complexes and based on the development of the synthesis of the new highly extended ligands containing fused TTF with benzocyano and pyrazine moieties [3,4], we decided to explore this new project. The dithiolene ligand precursor for preparing bisdithiolene complexes with highly extended ligands incorporating fused tetrathiafulvalene and pyrazine moieties, pyrazinedipropionitriletetrathiafulvalene, (pzdc-TTF) and their corresponding nickel, palladium and platinium bisdithiolene complexes were synthesized and characterized. These complexes, initially obtained under strictly anaerobic conditions all of them as diamagnetic species, nickel dianionic $TBA_2[Ni(pztdt)_2]$, palladium dianionic $TBA_2[Pd(pztdt)_2]$ and platinum dianionic $TBA_2[Pt(pztdt)_2]$, pztdt=pyrazinetetrathiafulvalenedithiolate.[5] These dianionic complexes are easily oxidised just by air exposure to their neutral states which present an extended range of stability. The exploration of these complexes to achieve more complex structures upon coordination to other metals through the pyrazine groups is currently under work.

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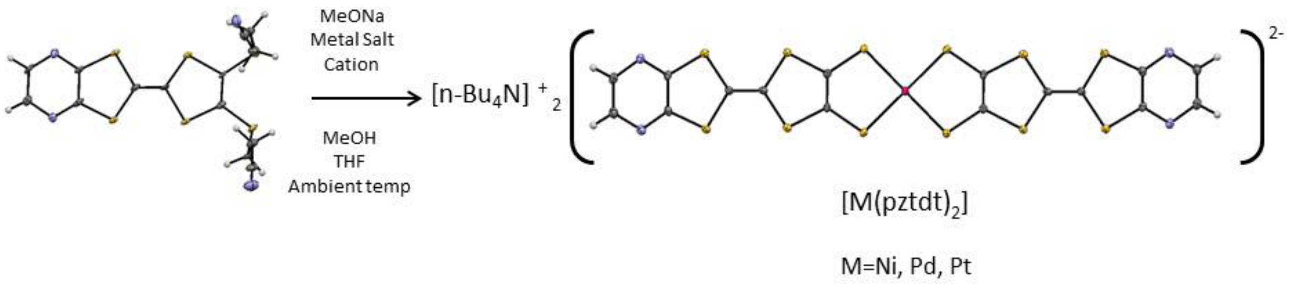

Scheme 1: Synthesis of the complexes $[n\text{-}Bu_4N]_2[M(pztdt)_2]$, M=Ni, Pd and Pt

PII-27

New charge-transfer salts based on π-conjugated donor molecules

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Organic charge-transfer salts are fascinating materials, with a large variety of physical ground states, ranging from insulators to semiconductors, metals and superconductors [1]. For organic electronics, π-conjugated molecules attracted attention as n- or p-type semiconductors. For correlated electron systems, materials are highly tunable by modifying the organic molecules utilizing the enormous flexibility of organic chemistry. Here, we present charge-transfer complexes with π-conjugated molecules representing either the donor or acceptor of these complexes. Besides known charge-transfer salts based on phenanthrene and coronene, we were able to synthesize charge-transfer salts with a new donor, DTBDT (dithieno-benzo-dithiophene) [2] as well as novel materials with picene. The complexes were grown by horizontal vapor growth technique or by crystal growth from solution. In this contribution we present the determined crystal structures, electrical transport measurements and spectroscopical investigations. These new materials crystallize in a so-called mixed stack configuration and are therefore possible candidates for a pressure or temperature induced ferroelectric transition. In addition to the experimental data, we present band-structure calculations, which were performed by density functional methods using the determined crystal structures as starting point. The interpretation of both is one approach to understand growing conditions of different stack configurations and is an important step towards the design of new charge-transfer complexes.

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TTF based multifuncontional Ligands and their corresponding electroactive metal complexes

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We report the synthesis, crystal chemistry and some physicochemical properties of multifunctional ligands and their corresponding electroactive metal complexes. These ligands are based on an association, directly or via a spacer, of tetrathiafulvalene as an electron-rich and redox active unit and on N-heterocycles as chelating fragment. These ligands were prepared in order:

1. To study and to evidence the existence of an intramolecular charge transfer band between the TTF (donor) and the acceptor units.
2. To prepare transition metal complexes with original structural and electronic properties and to study the influence of the metal cations on these properties.
3. To be used as potential precursors for the preparation of radical cation salts by the use of elctrocristallisation technique and therefore to obtain (semi)conducting crystalline materials.

The preparation and the X-ray structures of some of the ligands and their corresponding metal complexes will be reported with an eye on how each unit is influencing the electronic properties of these molecules

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PII-29

Crystal Structure and Physical Properties of Single-component Molecular Conductor, [Zn(tmdt)$_2$] with Tetrahedral Geometry

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We previously reported the crystal structure of single-component molecular conductor, [Zn(tmdt)$_2$], determined by synchrotron radiation powder X-ray experiments[1]. Recently, we succeeded in preparing single crystals of [Zn(tmdt)$_2$], then the crystal structures and physical properties were re-examined. Unlike typical single-component molecular metals M(tmdt)$_2$ (M = Ni, Pt, Au, Cu) with a planner coordination geometry, X-ray diffraction experiments revised that the molecular geometry of Zn(tmdt)$_2$ is tetrahedral, with the dihedral angle was 84.9°. As shown in Figure, the tmdt ligand on the right side of one Zn(tmdt)$_2$ molecule is stacked face-to-face with the ligand on the left side of the neighboring Zn(tmdt)$_2$ molecule to form a "tmdt dimer", with an arrangement closely resembling the two-dimensional molecular arrangement in the well-known "κ-type organic superconductors". Two-probe resistivity measurements were performed on single crystals of [Zn(tmdt)$_2$] with a size of approximately 70 µm. The room-temperature conductivity was 0.1 S/cm which was much smaller than those of planner [M(tmdt)$_2$] systems. The temperature dependence of the resistivity showed a semiconducting behavior with activation energy of 82 meV. NMR spin lattice relaxation rate suggested that [Zn(tmdt)$_2$] is a Mott insulator with a Mott gap of about 1400 K given by S-T model calculation, and forms a spin-singlet owing to the „tmdt dimer" structure[2]. Furthermore, both the magnetic and spin susceptibilities of [Zn(tmdt)$_2$] exhibited a minimum around 200 K. The susceptibilities were well fitted with the summation of a Curie-Weiss part and an S-T model part, which were consistent with NMR measurements.

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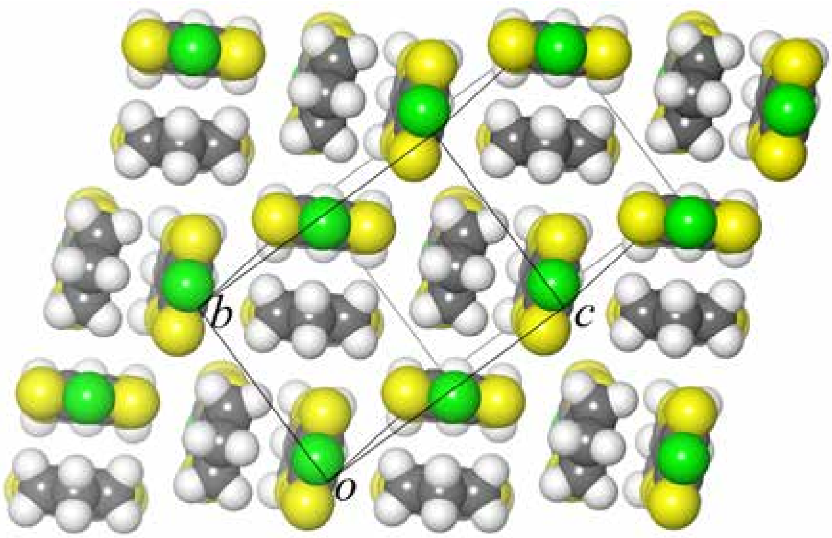

Figure: κ-type molecular arrangement" of [Zn(tmdt)$_2$]

PII-30

Chiral conductors from BEDT-TTF and novel chiral donor molecules

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More than 20 years ago Dunitz, Karrer and Wallis raised the question as to how the chirality of a material might affect its electrical properties, and prepared and studied enantiopure tetramethyl-BEDT-TTF donor molecules. More recently Rikken's studies on carbon nanotubes have shown that for a chiral conducting material in a magnetic field, the electrical resistance has a dependence on the handedness of the material. As part of our ongoing research in the synthesis of multifunctional conducting materials exhibiting chirality we report new families of enantiopure donor molecules and new salts incorporating chiral donors, anions or solvents. We have shown that it is possible to synthesise chiral molecular conductors using the readily available achiral BEDT-TTF donor molecule with inexpensive chiral electrolytes and racemic anions to bring about chiral induction, i.e. crystallisation of a chiral salt from a racemic precursor.

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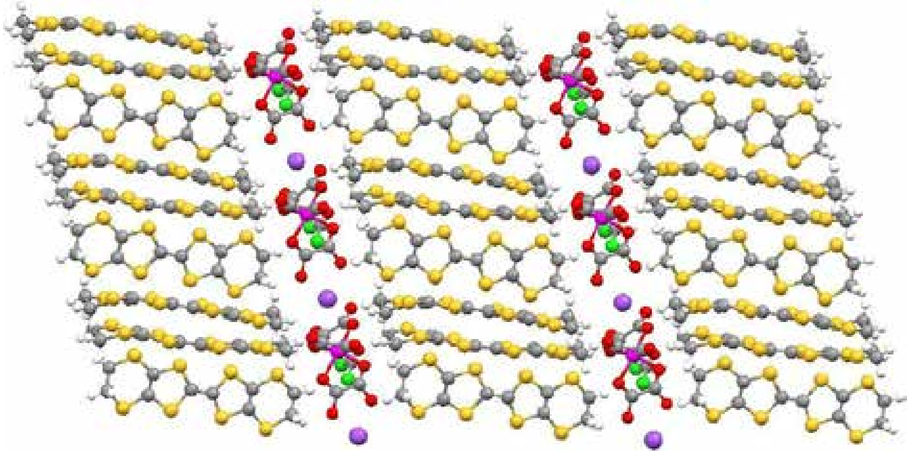

Figure: Layered packing of radical-cation salt with chiral anion

PII-31

Structure and dielectric properties of (naphthalene-1-aminium$^+$)(dibenzo[18]crown-6)[Ni(dmit)$_2$] showing pendulum motions

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Molecular motions in the crystal are attractive phenomena inducing the various physical properties such as dielectric response. We reported the crystal of (m-fluoroanilinium$^+$)(DB[18]crown-6)[Ni(dmit)$_2$] (DB[18]crown-6 = dibenzo[18]crown-6, dmit^{2-} = 1,3-dithol-4,5-dithiolate), exhibiting a ferroelectric transition at 346 K due to the dipole inversion caused by a flip-flop motion of m-fluoroanilinium$^+$ in the supramolecular rotator structure [1]. The other types of molecular motions such as pendulum motion in the solid state also may induce large dielectric responses. In the present work, we synthesized a crystal of (1NA)(DB[18]crown-6)[Ni(dmit)$_2$] (1) (1NA = naphthalene-1-aminium$^+$), in which pendulum motions of the extended p-electron system are expected Figure 1 shows the crystal structure of 1 at 300 K with the crystal system and space group of monoclinic and $P2_1/m$, respectively, having a mirror plane on the naphthalene ring. 1NA and DB[18]crown-6 molecules constructed a supramolecular cation structure, which formed an cationic layer in the ac plane. The cationic and anionic layers were alternately stacked along the b axis. The crystal structure at 93 K showed a symmetry reduction with the chiral space group of monoclinic, $P2_1$, where the mirror plane disappeared due to the inclination of the 1NA cation. A large dielectric response with the frequency dependence was observed in the temperature dependence of dielectric constant. The dielectric response may be related to the out of plane pendulum motion of the 1NA suggested by anisotropic thermal ellipsoids in the crystal structural analysis. The dielectric response will be discussed in terms of those of relaxer.

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Acknowledgement This work was supported by MEXT Grant-in-Aid for Scientific Research A and JSPS Core-to-Core Program, A. Advanced Research Networks

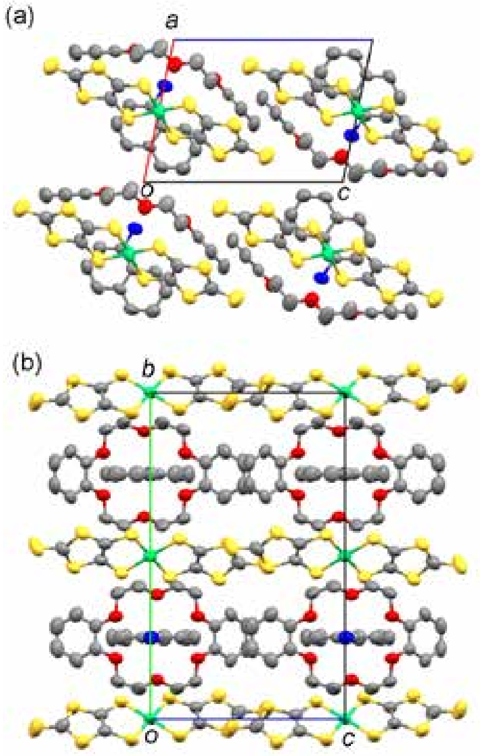

Figure 1: Crystal structure of 1

Self-assembled architectures with segregated donors and acceptor units of a dyad based on a monopyrrolo-annulated TTF-PTM radical derivative

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Organic molecules which contain electron donor (D) and electron acceptor (A) units linked by π-conjugated bridging groups (D-π-A dyads) are worthy of attention for the investigation of intramolecular electron transfer (IET) processes and in some cases for their associated bistability phenomena [1]. Recently, we have reported the D-A system based on a tetrathiafulvalene (TTF) electron π-donor connected to the polychlorotriphenylmethyl (PTM) radical electron acceptor by a vinylene bridge that exhibits bistability in solution through the application of external stimuli such as the polarity of the solvent or temperature [2-4]. Regarding the potential applications as electronic and optoelectronic devices, there is a high interest in obtaining TTF-based D-A systems that exhibit molecular packings with a complete segregation of the donor and acceptor fragments forming homostacks of the TTF subunits, since this is an important prerequisite for attaining electronic conductivities. Herein we report a new electron donor-acceptor dyad based on a PTM radical subunit linked to a TTF unit through a π-conjugated N-phenyl-pyrrole-vinylene bridge that has been synthesized and fully characterized (TTF-MP-PTM, Figure 1). The self-assembling ability of the radical dyad has been investigated by X-ray analysis showing supramolecular architectures with segregated donor and acceptor units where the TTF subunits are stacked forming 1-D chains. [5] Interestingly, preliminary experiments showed that this system exhibits conductivity upon the application of high pressures (with a resistivity of ca. 20 Ωcm at 6.5 GPa) and it has been corroborated by Raman spectroscopy.

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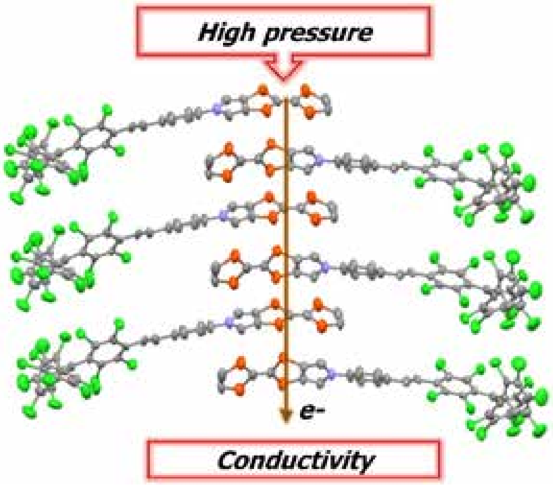

Figure 1: Molecular structure of TTF-MP-PTM exhibiting conductivity at high pressures

PII-33

The quest for soluble SCMM

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Among molecular conductors, Single Component Molecular Metals (SCMM) have been a topic of special interest because these materials offer the simplicity of being based on a unique neutral molecule, instead of the usual donor acceptor molecule couple. Our search for SCMM started with the preparation of [Au(a-tpdt)$_2$] (a-tpdt=2,3-thiophenedithiolate) neutral complex, characterised as a polycrystalline sample with a electrical conductivity of 7 S/cm, at room temperature [1]. Since then our efforts focused on the synthesis of similar transition metal complexes including ones with more extended dithiothiophene ligands [2], combining in the same molecule the “functionalities” of the two first examples of SCMM: a peripheral thiophene unit [1] and a TTF moiety [3]. Despite some successful attempts [4], possible technological applications are still limited by the poor solubility of SCMM, since their processing into suitable forms often requires single crystals or solution-based methods for depositing the complexes in a large area. Aiming at to overcoming this limitation, we focused our efforts in the preparation of thiophene-dithiolene transition metals complexes with ligands comprising alkyl groups. Indeed, this functionalization of the thiophenic moieties leads to more soluble neutral complexes [5]. In this communication we will present an overview of our results about SCMM based on transition metal bisdithiolenes with thiophenic units including recent results on alkyl substituted thiophenedithiolene complexes.

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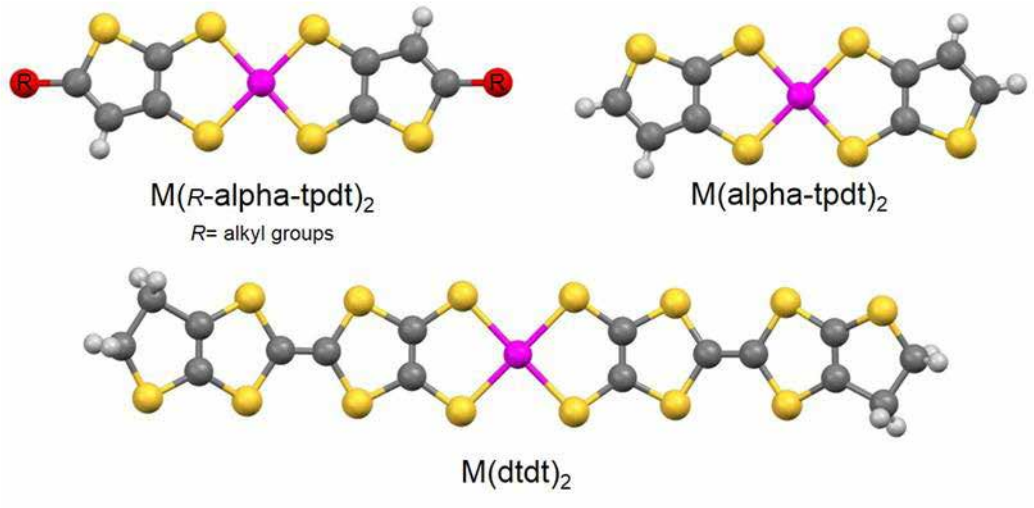

Figure 1: Examples of transition metals complexes with dithiothiophene ligands.

PII-34

Spin-Ladder structures and spin-ladder behaviour in thiophenic-TTF salts

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Since the first organic spin ladder reported, a dithiophene-tetrathiafulvalene salt ((DT-TTF)$_2$[Au(mnt)$_2$]) [1], different approaches have been made to enlarge the family of organic spin-ladders by subtle changes in donor and acceptor units [2]. Following this line of investigation we are able to report two new spin-ladder systems with the electron donor α-DT-TTF, the (α-DT-TTF)$_2$[Au(mnt)$_2$] and (α-DT-TTF)$_2$[Au(i-mnt)$_2$] [3,4]. Both compounds share the same donor ladder structure of the DT-TTF analogues [1,2]. In spite of the disorder in the thiophenic rings they present properties of a spin-ladder system with a room temperature conductivity of 2 S/cm and 0.1 S/cm, respectively [3,4]. The change of the gold complex [Au(mnt)$_2$] for its cobalt analogue, [Co(mnt)$_2$] leds to (α-DT-TTF)$_2$[Co(mnt)$_2$] with a new type of ladder structure. However it does not present magnetic spin-ladder behaviour due to the strong coupling between the disordered anion and donor chains [4].The superstructure of (DT-TTF)$_2$[Cu(mnt)$_2$] below the transition at 235 K was determined using synchrotron radiation and shed light into the spin carriers in this family of compounds. The low temperature structure of this compound presents a charge ordering scheme of alternating neutral (diamagnetic) and ionic (paramagnetic) donor units along the stacking axis which can explain why the magnetic interactions along the rails are smaller than along the rungs of the ladder [4].

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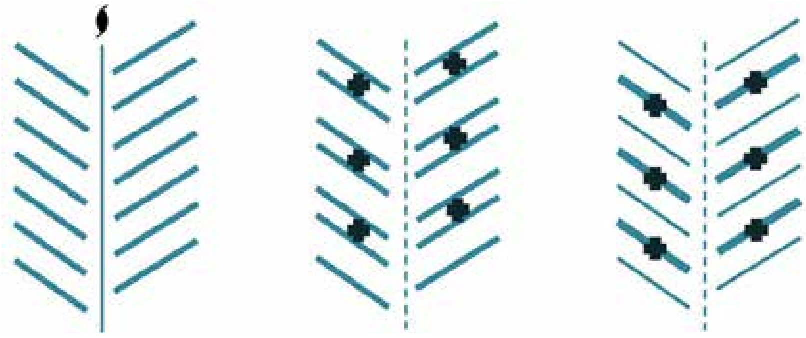

Figure 1: Representation of a pair of donor stacks connected by a screw axis (left) and possible schemes leading to a doubling of the staking axis parameter at low temperatures: charge ordering (center) and bond ordering (right).

PII-35

(CNB-EDT-TTF)$_n$X: Charge transfer salts based on a Dissymmetrical TTF donor with Cyano Coordinating Groups

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The chemical modification of the electron donor tetrathiafulvalene (TTF) has been extensively explored in search for new building blocks suitable for electroactive molecular materials, namely organic conductors and superconductores. We focused our interest on TTF-derivatives, with cyano units which can be used to coordinate transition metals by N atoms and to form in the solid state segregated and partially oxidized structures. In this context we have synthesized the dissymmetrical ET derivative cyanobenzene-ethylenedithio-tetrathiafulvalene (CNB-EDT-TTF) [1] and prepared by electrocrystallisation charge transfer salts of this donor with several anions X of different shapes and sizes. Their preparation and characterization by single crystal X-ray diffraction, electron transport measurements and band structure calculations are reported in this communication. With X= [M(mnt)$_2$]-, (M=Au, Ni) are obtained salts (CNB-EDT-TTF)$_2$X presenting a semiconducting behaviour with lower conductivity and high activation energy as expected from arrangement of the donors as chains of dimers. With small tetrahedral, octahedral and linear anions such as ClO$_{4-}$, PF$_{6-}$, and I$_{3,-}$ are obtained salts with another stoichiometry, (CNB-EDT-TTF)4X. These salts are a new type of 2D molecular metals based on a bilayer structure of the donors. The bilayer structure is induced by effective head to head interactions through the nitrile groups.

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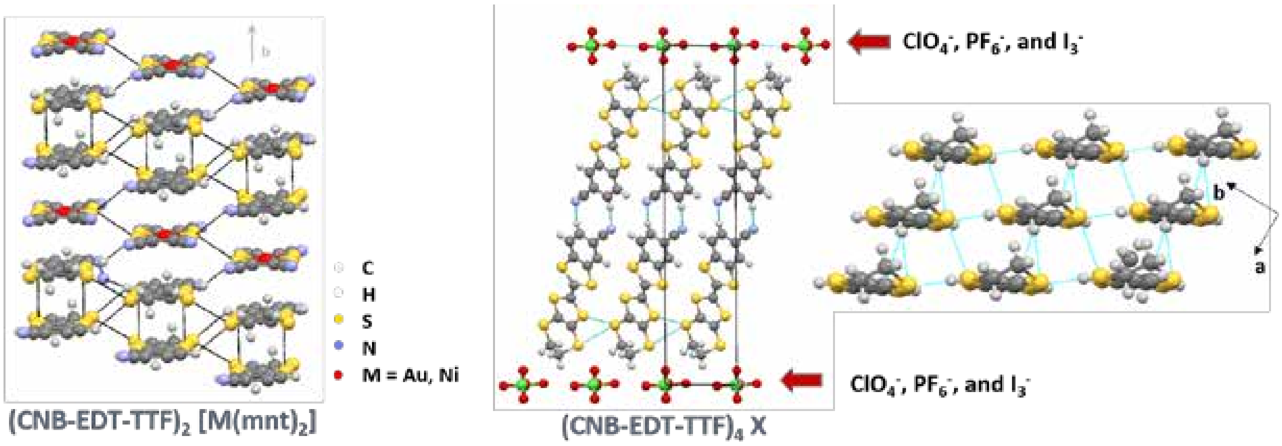

(CNB-EDT-TTF)$_2$ [M(mnt)$_2$]

(CNB-EDT-TTF)$_4$ X

Figure 1: Crystal structure of (CNB-EDT-TTF)$_2$ [M(mnt)$_2$](left) and (CNB-EDT-TTF)$_4$X (right).

PII-36

New radical cation salts of BEDT-TTF with $[ReCl_4(C_2O_4)]_2$- anion

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The design of hybrid materials combining conducting and magnetic properties in the same crystal lattice is one of the most developing trend in the field of molecular organic conductors. Here we present the synthesis and characterization of two new radical cation salts of the donor bis(ethylenedithio) tetrathiafulvalehe (BEDT-TTF) with paramagnetic tetrachloro(oxalate)rhenate(IV) $[ReCl_4(C_2O_4)]^{2-}$ anion. Up to now only two radical cation salts with this anion were known: (BEDT-TTF)$[ReCl_4(C_2O_4)]$ and θ-$(BEDT\text{-}TTF)_4[ReCl_4(C_2O_4)]\cdot C_6H_5CN$ [1] Electrocrystallization of BEDT-TTF in the presence of $[ReCl_4(C_2O_4)]^{2-}$ anion in different solvents at distinct temperatures and ratios of starting reagents afforded two new salts: β''-$(BEDT\text{-}TTF)_4[ReCl_4(C_2O_4)]$·Solv. (I) and α´-$(BEDT\text{-}TTF)_4[ReCl_4(C2O_4)]$·Solv. (II). The first salt was obtained when the mixture of solvents (C_6H_5CN+10 % C_2H_5OH) at room temperature was used. Using 1,1,2- trichloroethane $C_2H_3Cl_3$ as solvent is led to growing crystals II. New β''-salt is polymorphous modification of radical cation salt θ-$(BEDT\text{-}TTF)_4[ReCl_4(C_2O_4)]\cdot C_6H_5CN$. Compounds I and II have a layered structure, in which the conductive layers of BEDT-TTF alternate with anionic paramagnetic layers containing discrete anions $[Re(C_2O_4)Cl_4]^{2-}$ and guest molecules of crystallization solvent, fig. 1. Compound I has room-temperature conductivity of 0.4 S cm^{-1} and metalic behavior from 250 to 50 K. Below 50 K, salt becomes semiconducting. Salt II is a semiconductor.

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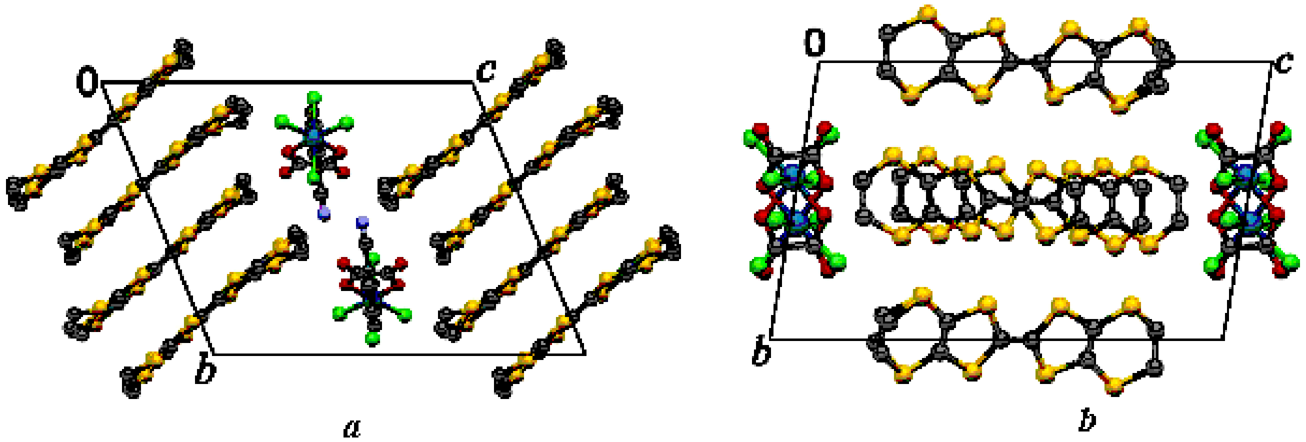

Figure 1: Crystal structure of β''-BEDT-$TTF_4[ReCl_4(C_2O_4)]$□Solv (a) and α´-$(BEDT\text{-}TTF)_4ReCl_4(C_2O_4)$□Solv (b)

A structural specificity of radical cation salts based on BEDT-TTF with $[ReX_6]^{2-}$ (X=Cl or Br) anion

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[2]*Institute of Problems of Chemical Physics RAS, Chernogolovka, Russia*

The synthesis, crystal and electronic structures of new radical cation salts based on the organic donor bis(ethylenedithio)tetrathiafulvalene (BEDT-TTF or ET) with the octahedral anions $[ReCl_6]^{2-}$ and $[ReBr_6]^{2-}$ are reported. δ-$ET_4[ReCl_6]$Solv (1), δ-$ET_4[ReBr_6]$Solv (2) and α'-$ET_4[ReBr_6]$Solv (3) were obtained by electrocrystallization with using BN, EtOH and DMF as reaction solvents [1]. All the salts (1-3) have layered structures. The 1 and 2 crystals are found to be isostructural and considerably different from early reported δ-$ET_4[ReCl_6]_{2-x}(CB)_x$ [2]. A summary of all known radical cation salts based on ET with octahedral complex anions of 5d-metals is given in the presentation. All crystals are semiconductors, their crystal structures show good example of molecular design. Phase transitions and superstructure in some of these crystals were found. Relationship between crystal structure and conducting properties is also discussed.

The study was supported by RFBR 14-03-31818 and Program of the Presidium of Russian Academy of Science.

[1] Unit cell parameters are: a=15.58, b=13.29, c=36.13Å, β=92.5˚, V=7475Å3 for (1); a=15.54, b=13.27, c=36.56 Å, β=91.5˚, V=7537Å3 for (2) and a=11.25, b=17.40, c=20.08 Å, α=70.3, β=79.8, γ=78.2˚, V=3597Å3 for (3).

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Combining chirality and conductivity in radical-cation salts using novel chiral donor molecules and chiral anions

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Field of Study: Synthesis of new molecules and molecular materials Combining Chirality and Conductivity in radical-cation salts with novel chiral donor molecules and chiral anions Jordan LOPEZ[a], Lee MARTIN[a], John WALLIS[a] and Hiroki AKUTSU[b] [a]Department of Chemistry, School of Science and Technology, Nottingham Trent University, Clifton Lane, Nottingham, NG11 8NS, United Kingdom. [b]Department of Chemistry, Graduate School of Science, Osaka University, 1-1 Machikaneyama-cho, Toyonaka, Osaka 560-0043, Japan. The organosulphur donors TTF and BEDT-TTF have produced a large number of radical-cation salts combining a variety of electrical properties, ranging from semiconductors to paramagnetic superconductors. The TTF based molecular conductors provide an excellent opportunity to explore the combination of chirality and conductivity in the same crystal lattice, which is of importance in understanding magnetochiral anisotropy as seen in carbon nanotubes.[1] In order to explore the effect of handedness on a conducting material a number of approaches have been taken to introduce chirality into TTF based conductors. Chiral radical-cation salts have included a variety of chiral anions, chiral donors, or have been electrocrystallised from a chiral solvent.[2-4] Using novel chiral donor molecules and chiral anions we have now synthesised and characterised several new families of radical-cation salts. Resistivity, magnetic and Raman data will be presented.

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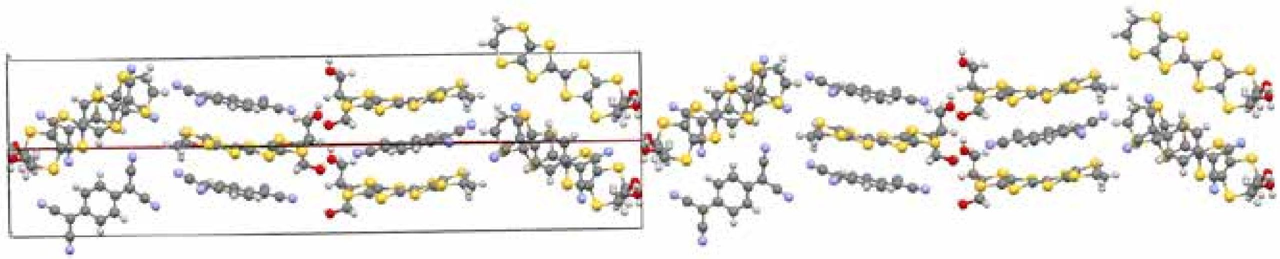

Chiral Donor TCNQ: Radical Cation Salt containing novel chiral donor molecule

PII-39

Topological property of exotic Dirac electrons in organic conductor [Pd(dddt)$_2$]

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The single-component molecular conductor [Pd(dddt)$_2$] (dddt = 1,4-dithiin-2,3-dithiolate) which displays exotic Dirac electrons under pressure has been found and studied using a tight-binding model consisting of four molecules per unit cell with HOMO and LUMO energy levels [1]. The model describes the following feature. The transition from the insulator to the zero-gap state (ZGS) with increasing pressure, is associated with an overlap between the convex HOMO band and the downward-convex LUMO band. The HOMO-LUMO interactions play an important role in the Dirac cone formation. In the present paper, we demonstrate a topological property to verify the exotic Dirac electron which exists as a pair of Dirac cones between the neighboring energy bands. The variation of the transfer energy with respect to the pressure, is estimated from an interpolation formula between the ambient pressure and the high pressure. The insulating state is a novel state which occurs as a merging of a pair of the Dirac electrons at the Gamma point. Further the ZGS is obtained by a pair of Dirac electrons which is located between the HOMO and LUMO bands. We calculate the parity of the wave function at the time reversal invariant momentum (TRIM), which is associated with the topological property originated from the inversion symmetry of the crystal [2] . It is shown that the product of all the parity of the wave functions below the Fermi level becomes negative in the presence of the Dirac electrons while it becomes positive in the absence of the Dirac electrons. Such a property is compared with that of organic conductor alpha-(BEDT-TTF)$_2$I$_3$, which has been well known as the two-dimensional Dirac electron system with the ZGS [3].

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PII-40

Multi-orbital kinetic effects on charge ordering of frustrated electrons on the triangular lattice

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Strongly correlated electron systems are often characterized by complex phase diagrams, reflecting an intricate interplay between magnetic, charge and orbital degrees of freedom. The role of the multi-orbital effects on the emergence of electronic orders on the frustrated triangular lattice at half-filling is investigated through a spinless extended Hubbard model. The latter is expected to describe layered triangular compounds, such as $AgNiO_2$ which exhibits a threefold charge order[1], or transition metal dichalcogenides where a variety of charge ordered phases have been observed[2]. Using complementary approaches, exact diagonalizations and unrestricted Hartree-Fock calculations, we unravel a very rich phase diagram with a competition between homogeneous, charge ordered and orbitally ordered phases, controlled by the strength of both local and short-range Coulomb interactions and by the hopping integrals of e_g orbitals. Among the unconventional electronic states, we will focus this presentation on the charge ordered phases. We find in particular, an original pinball liquid, which is a metallic charge ordered state where localized (pins) and itinerant electrons (balls) coexist, that was first devised in the context of quarter-filled organic conductors[3]. By changing the anisotropic ratio t'/t of the hopping integrals, an inverse pinball liquid occurs, where the role of charge-rich and charge-poor sites are interchanged, an insulating large unit cell $\sqrt{12} \times \sqrt{12}$ droplet phase can be stabilized[4], with a periodicity close to the $\sqrt{13} \times \sqrt{13}$ David Star charge order presents in $1T\text{-}TaS_2$. These electronic states are generic for the half-filled triangular lattice and are also found in the isotropic limit, which corresponds to the single-band spinful extended Hubbard model.

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PII-41

Ultrafast photoinduced phase conversion in two-dimensional molecular systems

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Recently, it has been experimentally claimed that an ultrafast photoinduced charge-order (CO) melting manifests itself with the time interval of about 25 fs just after the photoexcitation in a quasi-dimensional molecular crystal [1]. Motivated by this discovery, we theoretically investigate the ground- and excited-state properties of a two-dimensional CO system, using a spinless Fermion model and treating it by exact diagonalization [2]. Here, we consider both the nearest-neighbor (V) and next nearest-neighbor (V') intersite repulsions to include frustration. As the main result, we demonstrate that multi-electron excitations by one photon substantially occur as a result of the frustrating effect. These multi-electron excitations are naturally regarded as domain excitations, which are that a spatial region of the system lattice is excited simultaneously. In Fig. 1 (a), a calculated spectrrum of optical conductivity ($\sigma(\omega)$) is displayed. Here, the checkerboard-type CO ground state is assumed as the initial state. What is worth attention is that the number of excited electrons (N_{ex}) substantially exceeds one in the hatched regions, which indicates the possibility of multi-electron excitations by the photons with such energies. Furthermore, by the analysis of charge correlations, we find that such domain excitations not only suppress the original charge-ordering phase strongly (Fig. 1(b)) but also enhance another phase (Fig. 1(c)). As a result of such global change, the overall photoinduced optical conductivity spectra are also modified drastically from the original spectrum, showing midgap and metallic features.

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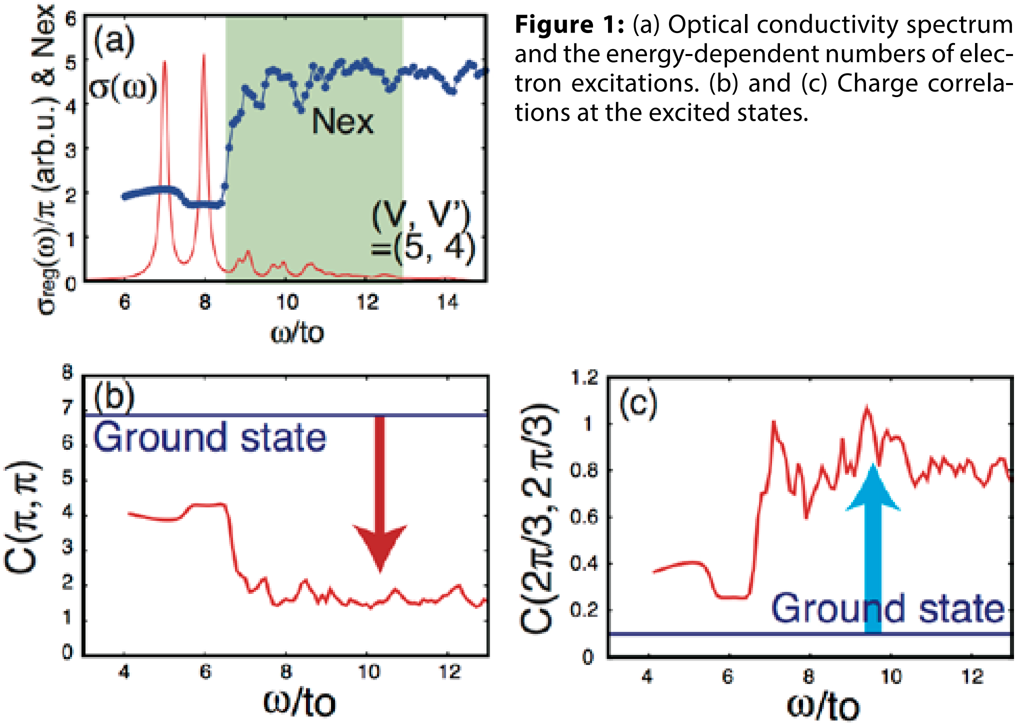

Figure 1: (a) Optical conductivity spectrum and the energy-dependent numbers of electron excitations. (b) and (c) Charge correlations at the excited states.

PII-42

Finite-temperature DMRG computation of NMR spin-lattice relaxation rate for spin-1/2 chains

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Low-dimensional quantum systems have been the subject of intense research for many decades. Powerful theoretical and numerical techniques, together with progresses in chemistry and experimental technology, give the possibility to perform a detailed study of real compounds. In this context the experimentally measured NMR (Nuclear Magnetic Resonance) spin-lattice relaxation rate $1/T_1$ represents a good challenge for theory. After having defined this quantity, and explained which information can be extracted from its measurement, I will show how recent developments in DMRG offer now the possibility to compute it. I will present some recent DMRG results for the NMR $1/T_1$ for spin-1/2 chains as a function of the temperature. The computed data show an excellent agreement with bosonization results available at low T. At finite T, regime hardly accessible via theoretical calculations, non-trivial behaviors are observed in some cases.

Stability of fractionally charged soliton and nonuniqueness of multimode Peierls transition

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In the first part, the stability of fractionally charged solitons in the “1100” background is discussed. The total energy of two solitons is numerically estimated, using the variational Monte Carlo method based on the matrix product state. The energy decreases as a function of the inter-soliton distance, which indicates that the effective force acting on the solitons is repulsive. Thus the fractionally charged soliton is stable. The relevance of the present results to $(DCNQI)_2X$ compounds will be discussed.[1] In the second part, multimode Peierls transition in the bond-centered square lattice is discussed. The frequencies of normal-mode phonons are numerically calculated in the tight-binding model with the electron-phonon interaction of the Holstein type. Simultaneous softening of multiple phonons occurs at a transition temperature. The free-energy calculation indicates that the transition is of the first order. The multimode Peierls states are highly degenerate. The results in the Holstein model will be compared with those in the SSH model.[2]

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PII-44

Pinball charge orderings driven by Coulomb interactions on triangular lattices

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Materials with competing electronic interactions on triangular lattices are a fertile ground for novel phenomena and original quantum phases, such as the spin liquid behavior observed in organic (κ-(BEDT-TTF)$_2$ Cu$_2$ (CN)$_3$,Me$_3$ EtSb[Pd(dmit)$_2$]$_2$) and inorganic (Cs$_2$ CuCl$_4$) quasi-two-dimensional materials. Geometrical frustration can play a similar role in charge ordering phenomena leading to poorly understood unconventional metallic and superconducting states. Remarkable examples are found in the quarter-filled organic salts, the layered cobaltates and the transition metal dichalcogenide 1T-TaS2. Motivated by the two-dimensional organic (theta-(BEDT-TTF)$_2$X) and inorganic (AgNiO$_2$) compounds, both with a triangular planar lattice structure but different bands and electronic fillings, we have analyzed the rich interplay between magnetic, orbital and charge degrees of freedom exhibiting exotic symmetry broken phases. By considering a multiband microscopic model which takes explicit account of electronic correlations, we have demonstrated that the emergence of charge ordered phases with unconventional metallic properties is a natural outcome in frustrated triangular geometries at various doping. In particular, we have shown by emplying complementary technics (DMFT, ED, UHF) that the competition between the frustration and the interactions in these systems lead to a robust pinball liquid phase, a quantum phase where the charges spontaneously separate into coexisting localized (pins) exhibiting Mott physics and band-like itinerant electrons (balls) moving on a honeycomb lattice. Interestingly, in the 1/4 filled, emergent heavy fermion behavior is observed at the Wigner-Mott transition [1] while at 1/2 filling, such peculiar state of matter can be stabilized thanks to Hund's couplings [2]. We argue that the charge ordered metallic phase of AgNiO2 is a neat experimental realization of such pinball liquid, which explains many of the observed experimental features.

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PII-45

PII-45

Intramolecular charge-transfer: Hubbard model parameters from quantum-chemical calculations

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Mixed-stack charge transfer (MS-CT) crystals contain one-dimensional stacks of planar π-electron donors (D) and acceptors (A) arranged face-to-face with a sizable overlap of frontier orbitals, resulting in delocalized electrons and fractional charges $\ldots D^{\rho+}A^{\rho-}D^{\rho+}A^{\rho-}\ldots$ MS-CT crystals are an interesting example of multifunctional molecular materials, and are currently investigated as promising all-organic ferroelectrics and potential multiferroics. Their rich phase diagram provides opportunities to study quantum phase transitions, multistability and photoinduced phase transitions. The standard microscopic model to describe this family of material is the Modified Hubbard model accounting for electron-phonon coupling (Peierls coupling), electron-molecular vibrations coupling (Holstein coupling) and electrostatic interactions. Traditionally, model parameters are derived from experiments; [1] but recently an approach was proposed to extract model parameters from quantum-chemical calculations. [2] Here we present the results of an extensive set of calculations aiming to the parametrization of a large family of MS-CT salts in order to shed light on the complex physics governing these promising multifunctional materials.

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PII-46

Intramolecular charge-transfer: Model parameters from quantum-chemical calculations

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Molecules that can be switched between two or more states through the application of an external stimulus (i.e., light, electric field, solvent) are the basis for multistable materials. D-π-A dyads, where an electron donor group (D) is bound to an electron acceptor (A) through a π-conjugated bridge, are particularly interesting in this respect. Intramolecular electron transfer (IET) governs the low energy physics of D-π-A dyads and is well described by essential state models, a family of parametric Hamiltonian, usually parametrized against experimental data. Recently new materials have been proposed, [1,2] based on TTF (tetrathiafulvalene) donor group, where both intra and intermolecular electron-transfer play a role. To build reliable models for these intriguing materials and even more to guide the design of materials with requested properties we propose new strategies to parametrize essentials state models from quantum-chemical calculations. Two different approaches will be presented: one based on the analysis of low-energy excited states of the solvated molecules through (TD)-DFT calculations of the ground and low-lying excitations of TTF-PTM dyads in solution. A second strategy only relies on ground state properties and specifically on DFT (UCAMB3LYP/6-31+G*) and PM7 calculation of the molecular dipole moment in an applied electric field.

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PII-47

Pseudo-Orbital approach to spin-orbit coupling in organics

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Spin-orbit coupling (SOC) has recently come to the forefront of materials research, due to its potential role in stabilizing new and exotic magnetic and electronic phases [1]. In inorganic materials based on heavy d-, f-, and p-block elements, the natural energy scale of spin-orbit coupling may be estimated from the atomic SOC constants, and may be competitive with the kinetic (hopping) and potential (Coulomb repulsion) energy. In this case, it is often possible to confine the treatment of SOC to a few orbitals such as the highest occupied p- or d-orbitals, or a subset thereof. The inclusion of such effects in insulators, for example, leads to strongly anisotropic interactions between local spin-orbital coupled moments [2]. For organic materials, the existence of potentially hundreds of relevant molecular orbitals significantly complicates this picture. In this contribution, we discuss how SOC can be considered within a manifold including three auxiliary pseudo-orbitals associated with each partially occupied orbital in the ground state. Using this approach, the orientation and magnitude of anisotropic exchange may be easily predicted, as well as estimated via DFT calculations [3,4]. The results of this approach for a range of sulfur and selenium based organic radicals and charge transfer salts will be discussed. A particularly important result is that the effective magnitude of SOC is generally orbital dependent in organics, allowing potential for tuning at the chemical level [5].

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BEDT-TTF charge estimation revisited: a quantitative crystallographic approach

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A correct estimation of the partial positive charge localized on the BEDT-TTF (ET) molecule in the conductive salts is of paramount importance in many practical cases.[1] A technique accounting variations of all bond lengths inside the central TTF core was a step forward from earlier attempts to determine ET molecular mean charge solely by a variation of the central C=C bond length.[2] Unfortunately, this technique provides reliable results only in the cases of perfectly refined structures (R1 < 5%) that seriously restricts its applicability as many of single crystals of ET salts demonstrating interesting physical properties possess essential sources lowering quality of the crystal structure (twinning, intrinsic disorder, superstructures, high mosaicity, etc.). Moreover, even in the cases of low R1 structures some hidden errors persist. The errors stem from the peculiarity of ET molecular structure where light carbon (C) atoms of TTF core are surrounded by heavy sulfur (S) atoms.[3] While the coordinates of S atoms usually are reliably solved even in the high R1 structures, C atoms, screened by heavier S ones, often take apparently unreasonable coordinates. These errors in the C atoms coordinates make C=C bond lengths unwelcomed in the accurate procedure of ET charge estimation. Thus, I present a new formula based on a thorough analysis of 895 structures of ET salts found in the latest release of CCDC database (v. 5.36, Nov 2014). The formula takes mutual coordinates of heavier sulfur atoms as a statistically reliable basis for the estimation of ET charge. Applicability of the formula was tested on the important examples of ET salts with phase transitions accompanied by charge ordering.

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08:00		Opening					08:00
09:00		Session 1: Superconductivity I	Session 5: Dirac electron systems	Session 9: Charge Order II	Session 12: Quantum spin liquids II	Session 16: Ultrafast Phenomena	09:00
10:00		Coffee Break	Coffee Break	Coffee Break	Coffee Break	Coffee Break	10:00
11:00		Session 2: Design of new materials I	Session 6: Charge Order I	Session 10: Superconductivity II	Session 13: Ferroelectricity II	Session 17: Functional Materials - Intramolecular Properties	11:00
12:00		Lunch	Lunch	Lunch	Lunch	Closing	12:00
13:00				Session 11: Ferroelectricity I			13:00
14:00		Session 3: Functional materials	Session 7: Quantum spin liquids I		Session 14: Design of new Materials II		14:00
15:00	Check-In		Coffee Break	Excursion	Coffee Break		15:00
16:00		Coffee Break Session 4: Mott physics I	Session 8: Magnetic field effects		Session 15: Mott Physics II		16:00
17:00							17:00
18:00							18:00
19:00	Dinner Welcome Reception	Dinner	Dinner		Dinner		19:00
20:00		Advisory Board Meeting	Poster Session I		Poster Session II		20:00
21:00							21:00
22:00							22:00