International Workshop

Theory Meets Experiment in Low-Dimensional Structures with Correlated Electrons

Prague, July 1–4, 2019

Organizers

Ján Minár
New Technologies Research Centre, University of West Bohemia

Pavel Jelínek
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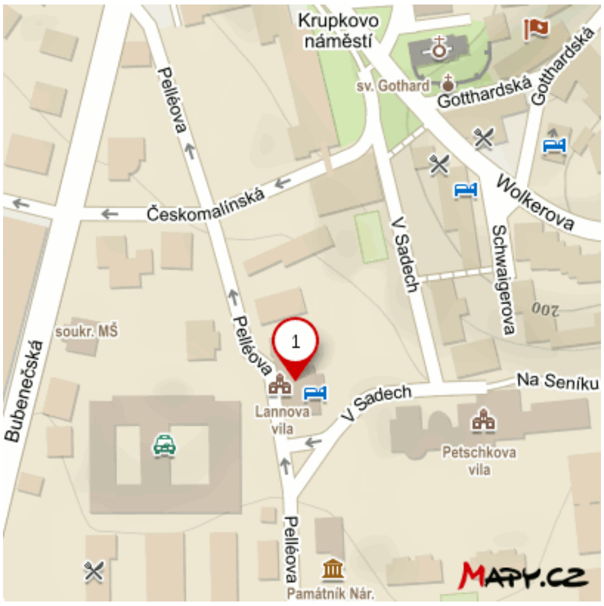
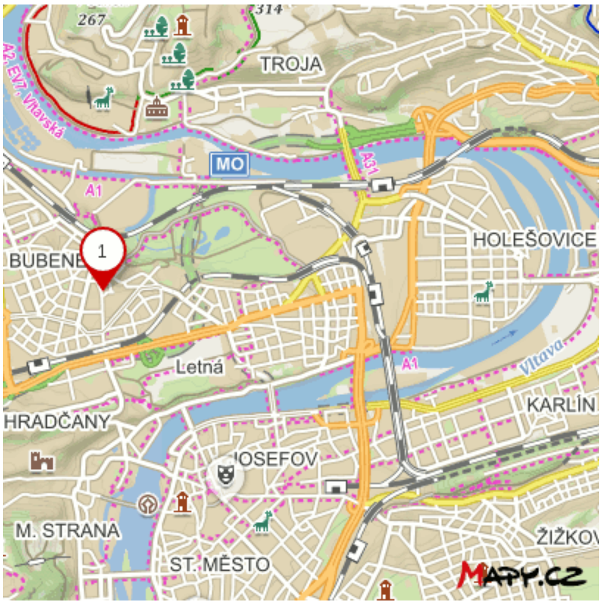
Institute of Physics, Czech Academy of Sciences

Workshop venue

Villa Lanna
V Sadech 1, Prague, Czech Republic





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Program

Monday, July 1		Tuesday, July 2		Wednesday, July 3		Thursday, July 4	
time	name	time	name	time	name	time	name
09:00	workshop opening	09:00	**F. Evers**	09:00	**H. Wende**	09:00	**J. Repp**
09:10	**H. Brune**	09:35	V. Pokorný	09:35	Ch. Wäckerlin	09:35	C. Bonizzoni
09:45	V. Bellini	09:55	A. Cahlík	09:55	A. Herman	09:55	J. Schusser
10:05	I. R. Reddy	10:15	coffee	10:15	coffee	10:15	coffee
10:25	coffee	10:45	**N. Lorente**	10:45	**I. Di Marco**	10:45	**J. Kuneš**
10:55	**A. Khajetoorians**	11:20	R. Ortiz	11:20	H. Ullah	11:20	G. Fratesi
11:30	S. W. DSouza	11:40	T. Novotný	11:40	S. Achilli	11:40	E. Tereshina-Chitrova
11:50	J. Chakraborty	12:00	lunch	12:00	lunch	12:00	lunch
12:10	lunch	14:00	**J. Fernández-Rossier**	14:00	**L. H. Tjeng**	14:00	**M. Verstraete**
14:10	**A. F. Otte**	14:35	M. Švec	14:35	O. Šipr	14:35	P. Kratzer
14:45	R. Korytár	14:55	L. Buimaga-Iarinca	14:55	A. Sadeghi	14:55	V. Kumar
15:05	**O. Maršálek**	15:15	coffee	15:15	coffee	15:15	coffee
15:40	coffee	15:45	**T. Wehling**	15:45	**A. Hariki**	15:45	**A. Shick**
16:10	**J. I. Pascual**	16:20	M. Pizzochero	16:20	M. Winder	16:20	H. Janssen
16:45	S. Pons	16:40	H. Ud Din	16:40	B. Chatterjee	16:40	F. Alarab
17:05	A. Kadlecová	17:00	J. Krempaský				

Invited contributions

Harald Brune (EPF Lausanne, Switzerland)
Zero-Dimensional Magnetic Structures with Correlated Electrons

Alexander Ako Khajetoorians (Radboud University, Nijmegen, Netherlands)
Unraveling the glassy nature of the magnetic state of Nd

Sander Otte (Delft University of Technology, Netherlands)
Quantum simulation through atomic assembly

Ondřej Maršálek (Charles University, Czech Republic)
Nuclear quantum effects in molecular systems through path integrals

J. I. Pascual (nanoGUNE, San Sebastian, Spain)
Mapping Yu-Shiba-Rusinov states in atomic and molecular impurities

Ferdinand Evers (U Regensburg, Germany)
Aspects of Topology and Quantum Interference in Single Molecule Transport

Nicolas Lorente (CSIC-EHU, San Sebastian, Spain)
Isospin-flip spectroscopy

Joaquín Fernández-Rossier (INL Braga, Portugal)
Spin correlations in graphene nanostructures probed with STM

Tim O. Wehling (U Bremen, Germany)
Interactions in superlattice quantum materials

Heiko Wende (U Duisburg-Essen, Germany)
Europium cyclooctatetraene nanowire carpets: a low-dimensional, organometallic ferromagnet

Igor Di Marco (APCTP, South Korea)

Ab-initio treatment of the Lanthanides: from solids to clusters and atoms

Liu Hao Tjeng (Max Planck - CPfS, Dresden, Germany)
Direct Imaging of Orbitals using Inelastic X-Ray Scattering

Atsushi Hariki (TU Wien, Austria)
LDA+DMFT approach to resonant inelastic x-ray scattering in correlated materials

Jascha Repp (U Regensburg, Germany)
Actuating and probing a single-molecule switch at femtosecond timescales

Jan Kuneš (TU Wien, Austria)
Towards predictive theory of two-particle spectroscopies in strongly correlated materials

Matthieu Verstraete (U Liège, Belgium)
Thermal displacements in microscopies and spectroscopies

Alexander Shick (Czech Academy of Sciences, Prague, Czech Republic)
More surprises in f-electron magnetism

Abstracts

Monday, July 1

Zero-Dimensional Magnetic Structures with Correlated Electrons

Harald Brune

Institute of Physics, EPFL, CH-1015 Lausanne, Switzerland

Zero-dimensional magnetic structures are single magnetic atoms. In order for them to have non-trivial magnetic properties that can be addressed and potentially used, they are adsorbed onto single crystal surfaces. The magnetic eigenstates of their electrons emerge from the crystal field of the adsorption site and from spin-orbit coupling, thus from electron correlations. The stability of the ground state depends on the excited states that can be reached by electron or phonon scattering. Also the nuclear spin states play an important role in the magnetic stability.

Single Ho atoms on MgO(100)/Ag(100) are the smallest and most stable magnets known today. We show recent results on their thermal and magnetic field stability. They yield an onset temperature for magnetization switching of 45 K and an estimated coercitive fields of 60 T. We show that Landau-Zener tunneling through avoided level crossings involving the nuclear spin states can be used to prepare the magnetic states by magnetic field sweeps that are by orders of magnitude smaller than the coercitive field.

Many rare-earth atoms adsorbed on graphene exhibit intense inelastic conductance steps in scanning tunneling spectroscopy. Their origin is an intra-atomic spin-excitation between the 4f and 6s spin-moments. For a given valency, the exchange energy between these moments is largely independent of the 4f element [H. S. Li et al., J. Phys.: Condens. Matter 3, 7277 (1991)] and therefore we observe a linear behavior of excitation energy with filling of 4f levels. Note that this excitation takes place on the 6s, and possibly also 5d, states and not on the 4f levels that are to localized to interact with the tunnel electrons. Due to charge transfer to graphene, the 6s spin moment is high. This leads to giant spin-polarization observed for Dy atoms on graphene grown on Ir(111). The first 4f adatom for which STM spin-contrast was reported is Ho/MgO, in that case the spin-contrast was only 2–4 pm in apparent height. Dy atoms on graphene exhibit up to 60 pm apparent height contrast between the up and down states. We attribute this to Dy transferring almost an entire electron to graphene [M. Pivetta et al., Phys. Rev. B 98, 115417 (2018)], leading to a half-filled 6s shell. Its spin-polarization is therefore close to 100 %, and the projection of the spin onto the out-of-plane axis is stable due to the stability of the Dy 4f states and the exchange coupling to them.

Graphene-mediated exchange coupling between Co single spins and Ni substrates

Valerio Bellini (1), Alessandro Barla (2), Carlo Carbone (2), Stefan Heinze (3)

(1) Istituto Nanoscienze - CNR Nano S3, via G. Campi 231/A, 41125 Modena, Italy, (2) Istituto di Struttura della Materia (ISM), Consiglio Nazionale delle Ricerche (CNR), I-34149 Trieste, Italy, (3) Institute of Theoretical Physics and Astrophysics, University of Kiel, D-24098 Kiel, Germany.

We present a density-functional theory study of the exchange coupling between molecular/single atom spin systems and a magnetic substrate across a graphene spacer. We concentrate on cobaltocene ($CoCp_2$) molecules and Co adatoms adsorbed on graphene decorated Ni(111) substrates. The role of graphene is to preserve the adsorbate magnetic moments, acting as an electronic decoupling layer, while allowing effective spin communication between the adsorbate and the substrate. In fact, sizable magnetic coupling is predicted by the theory, and in some fortunate cases can be also rationalized in term of spatial and energy matching between adsorbate and substrate orbitals.

This is the case of cobaltocene, where in virtue of the peculiar magnetic properties of the molecule, the coupling mechanism is more accessible, and the description of the employed theoretical methodology suffices to rationalize it, and even tailor it by, for instance, intercalation of different ferromagnetic metal monolayers, such as Fe and Co, between graphene and the Ni substrate [S. Marocchi et al., Phys. Rev. B. 88, 144407 (2013)].

Concerning single atom spin adsorbates, we first concentrate on Co adatoms, where a thorough experimental analysis by STM and XMCD experiments has been also conducted, and where a consistent description between theory and experiments can be found only by considering the presence of more adsorption sites characterized by different magnetic properties [A. Barla et al., ACS Nano 10, 1101 (2016)]. If we extend the analysis to the other elements of the 3d series, i.e. from Sc to Fe, although some trends across the series could be found [V. Bellini et al., to be submitted], no clear understanding of the magnetic coupling can be set forward. Moreover, the employed methodology fails in the comparison with XMCD experiments performed on V adatoms, sign that more refined multi-reference methods might be necessary to correctly describe the ground state magnetic properties of these systems.

Spin State Manipulation of Nickel Quinonoid by Chemisorbed to a Co(001) Substrate

Indukuru Ramesh Reddy (1), Peter M. Oppeneer (2), Kartick Tarafder (1)

(1) National Institute of Technology Karnataka, India, (2) Uppsala University, Sweden

We studied the structural, electronic and magnetic properties of recently synthesized Ni(II)-quinonoid molecule adsorbed on magnetic Co(001) substrate by means of density functional theory (DFT)+U calculations. A strong covalent interaction at the interface increases the Ni-O(N) bond lengths in the chemisorbed molecule leading to a spin state switching in the molecule from $S = 0$ state (in the gas phase) to $S \approx 1$ state upon adsorption on the Co(001) substrate. Our DFT+U calculations shows that the molecule is ferromagnetically coupled with substrate. The exchange mechanism between the Co surface and metal centre in the molecule (Ni atom) was carefully investigated. It has been found that an indirect exchange interaction via quinonid ligands of the molecule, stabilizes the spin moment in the molecule in ferromagnetic alignment with the Co surface magnetization.

Unraveling the glassy nature of the magnetic state of Nd

Alexander Ako Khajetoorians

Scanning Probe Microscopy, Institute for Molecules and Materials, Radboud University, The Netherlands

Spin glasses are a fascinating but poorly understood magnetic state of matter, appearing in complex alloys like iron-based superconductors and magnetically-doped metals, like Mn-doped Cu. Intricately linked to spin frustration and disorder, spin glasses exhibit no long-range magnetic order upon cooling. Likewise, spin glasses exude the distinguishing phenomenon known as aging, where the magnetic state is dependent on the history of the material, unlike quantum spin liquids. Here we show a new type of non-collinear spin glass order, or so-called spin-Q glass, observable in elemental Nd(0001) in the absence of disorder. Using spin-polarized scanning tunneling microscopy down to mK temperature, we visualize the competing local non-collinear order at the atomic-scale and probe the subsequent aging phenomena in response to variable magnetic field and temperature. We relate the resultant glassy behavior to the crystalline symmetry, which leads to competing magnetic interactions. Results from ab initio calculations will also be discussed, as well as applications of the spin Q glass concept to potential brain-inspired hardware.

Electronic Structure of antiferromagnetic Mn_2Au: A Novel spintronic compound

S. W. DSouza, J. Minár

New Technologies Research Centre, University of West Bohemia, Univerzitní 8,
CZ-306 14 Pilsen, Czech Republic.

Electronic structure of antiferromagnetic Mn_2Au has been studied by utilizing first-principles density-functional theory calculations based on the Green’s function technique. The total density of states reveals contributions from the Au 4f and two Mn 3d states with collinear antiferromagnetic (AF) order with localized magnetic moments. Our ground state band structure calculations representing the Bloch spectral functions shows that the essence of the AF-switching induced by Néel spin-orbit torques in Mn_2Au is in the ΓX direction of the Brillouin zone indicating a band crossing for Neel vector oriented along [100] direction. The one step model angle resolved photoemission calculations in a k_z band map for photon energies ranging from 400−1000 eV shows dispersive spectral weight at Fermi level along ΓΣ direction of the Brillouin zone.

Interplay of covalency, spin-orbit coupling, and geometric frustration in the $d^{3.5}$ system $Ba_3LiIr_2O_9$

Jayita Chakraborty

Indian Institute of Science Education and Research Bhopal, Bhauri, Bhopal, India

The interplay of strong spin-orbit coupling, electron electron correlation, and crystal-field splitting in 5d transition metal oxides have recently attracted much attention both theoretically and experimentally. Here we investigate the electronic and magnetic properties of $d^{3.5}$ iridate $Ba_3LiIr_2O_9$ using first-principles electronic structure calculations. The results of the calculations reveal that the system lies in an intermediate spin-orbit coupling (SOC) regime. There is strong covalency of Ir-5d and O-2p orbitals. SOC, together with covalency, conspires to reduce the magnetic moment at the Ir site. By calculating the hopping interactions and exchange interactions, it is found that there is strong antiferromagnetic intradimer coupling within an Ir_2O_9 unit and other antiferromagnetic interdimer interactions make the system frustrated. The anisotropic magnetic interactions are also calculated. The calculations reveal that the magnitude of the Dzyaloshinskii-Moriya (DM) interactions parameter is small for this system. The effects of small electron and hole doping on DM interactions are also investigated here. The magnetocrystalline anisotropy energy is significant for this system, and the easy axis lies on the ab plane.

Quantum simulation through atomic assembly

Sander Otte

Delft University of Technology, The Netherlands

The magnetic and electronic properties of materials often find their origin in basic atomic- scale interactions. Yet, due to the large number of atoms involved, many phenomena can be very difficult to predict: we call these ’emergent’. The ability to build structures atom-by-atom by means of scanning tunneling microscopy (STM) provides an excellent platform to explore emergence as a function of system size. By properly tuning the anisotropy and coupling of magnetic atoms on a thin insulator, we have been able to engineer finite spin chains hosting spin waves [1] as well as the beginnings of a quantum phase transition at a critical magnetic field [2]. In a more recent experiment, we have engineered spin structures that are frustrated by design, exhibiting a spin spiral that can snap between different configurations. Unfortunately, the maximum size of assembled structures is often limited due to e.g. crystal impurity and crystal strain. In this talk, I will present a way to mitigate these limitations and show recent advances in sample preparation that will allow us to build much larger spin structures [3].

[1] A. Spinelli et al., Nature Materials 13 (2014) 782.
[2] R. Toskovic et al., Nature Physics 12 (2016) 656.
[3] J. Gobeil et al., Surface Science 679 (2019) 202.

Signatures of a coherent ad-atom Kondo lattice

Richard Korytár

Charles University, Prague, Czech Republic

Metallic surfaces are an ideal playground for exploring correlation physics in magnetic chains made of ad-atoms or molecular networks. I will discuss manifestations of Kondo-lattice phenomenology in such chains and the possibility to observe heavy-fermion physics.

[1] M. Moro-Larages, R. Korytár, M. Piantek, R. Robles, N. Lorente, J. Pascual,
M. R. Ibarra and D. Serrate, Nature Communications, to appear, (2019).

Nuclear quantum effects in molecular systems through path integrals

Ondřej Maršálek

Charles University, Faculty of Mathematics and Physics, Prague, Czech Republic

Methodology based on the path integral formulation of quantum mechanics offers a computationally tractable way to include nuclear quantum effects in atomistic simulations of extended condensed-phase systems. Here, we will review some of the required theoretical and algorithmic ingredients, with focus on ab initio molecular dynamics simulations. While all static quantities can be calculated numerically exactly, dynamical information can be obtained from these simulations approximately within the framework of centroid or ring polymer molecular dynamics. Ring polymer contraction is a controlled approximation which allows for an efficient evaluation of physical interactions in a path integral system. It is especially useful with our recent extension to ab initio interactions, where it accelerates practical simulations by up to two orders of magnitude. Importantly, it can be combined with centroid and ring polymer molecular dynamics to obtain approximate dynamics from path integral simulations in a computationally efficient way. We will illustrate the use of this methodology with results of ab initio path integral molecular dynamics simulations of aqueous solutions and small molecular clusters at both ambient and low temperatures.

Mapping Yu-Shiba-Rusinov States in atomic and molecular impurities

Nacho Pascual, Javier Zaldivar, Carmen Rubio-Verdú, Deungjang Choi,
Stefano Trivini

CIC nanoGUNE, Tolosa Hiribidea 76, 20018 Donostia - San Sebastian, Spain

Magnetism and superconductivity are phenomena that cannot simultaneously exist in the same region of a material. At the atomic scale, magnetic impurities scatter Cooper pairs as a potential with broken time-reversal symmetry, and create bound states inside the superconducting gap, named Yu-Shiba-Rusinov (YSR) states [1]. We investigate the YSR states caused by several atomic and molecular systems, resolving their origin and exploring basic properties of BCS superconductors.

YSR states are localized around magnetic impurities and their amplitude distribution reflects intrinsic properties of the magnetic system and of its interaction with the superconductor. We will present recent results about the spatial distribution of YSR states on the surfaces of two conventional superconductors, Pb(111) and β-Bi_2Pd(100) [2,3]. While in the close proximity to the impurity, YSR states may reflect the projected shape of spin polarized orbitals of atomic adsorbate, on the long range, they picture scattering processes in the superconducting bands

[1] B. W. Heinrich, J. I. Pascual, and K. J. Franke, ArXiv 1705.03672, (2017).
[2] D. J. Choi, C. Rubio, J. Bruijckere, M. M. Ugeda, N. Lorente, J. I. Pascual, Nat. Comm. 8, 15175 (2017).
[3] D. Choi, C. García Fernandez, E. Herrera, C. Rubio-Verdu, M. Ugeda, I. Guillamon, H. Suderow, J. I. Pascual and N. Lorente, Phys. Rev. Lett. 120, 167001 (2018).

How small a superconductor can be?

S. Pons (1), S. Vlaic (1), T. Zhang (1), A. Assouline (1), A. Zimmers (1),
C. David (2), G. Rodary (2), J.-C. Girard (2), D. Roditchev (1) and H. Aubin (1,2)

(1) Laboratoire de Physique et d'Étude des Matériaux, Ecole de Physique et Chimie Industrielles, CNRS, Sorbonne Université, Paris, France, (2) Centre de Nanosciences et de Nanotechnologies, CNRS, Université Paris-Sud, Marcoussis, France

In 1959, physicist P. W. Anderson conjectured that superconductivity can exist only in objects where superconducting gap energy is larger than its electronic energy level spacing. In our work [1,2], we were able to address this limit in single nanoparticles of various sizes without suffering from inverse proximity effect. Here we report on a scanning tunneling spectroscopy study of superconducting lead nanocrystals grown on the (110) surface of InAs. The electronic transmission at the InAs/nanocrystal interface is weak; this leads to Coulomb blockade and enables the extraction of electron addition energy of the nanocrystals. The addition energy displays superconducting parity effect, a direct consequence of Cooper pairing. Due to the electrons forming Cooper pairs when an electron is added to the nanocrystal superconductor, the additional energy is different whether there is an even or odd number of electrons. Studying this parity effect as a function of nanocrystal volume, we find the suppression of Cooper pairing when the mean electronic level spacing overcomes the superconducting gap energy, thus demonstrating unambiguously the validity of the Anderson criterion.

[1] Superconducting parity effect across the Anderson limit, S. Vlaic, S. Pons, T. Zhang, A. Assouline, A. Zimmers, C. David, G. Rodary, J.-C. Girard, D. Roditchev, H. Aubin, Nature Comm. 8, 14549 (2017).
[2] Quantum confinement effects in Pb nanocrystals grown on InAs, T. Zhang, S. Vlaic, S. Pons, A. Assouline, A. Zimmers, D. Roditchev, H. Aubin and G. Allan, C. Delerue, C. David, G. Rodary, J.-C. Girard, Phys. Rev. B 97, 214514 (2018).

Quantum Phase Transitions in Superconducting Quantum Dots

Alžběta Kadlecová (1), Martin Žonda (1,2), Vladislav Pokorný (1),
Tomáš Novotný (1)

(1) Department of Condensed Matter Physics, Faculty of Mathematics and Physics, Charles University in Prague, Ke Karlovu 5, CZ-121 16 Praha 2, Czech Republic, (2) Institute of Physics, Albert Ludwig University of Freiburg, Hermann-Herder-Strasse 3, 791 04 Freiburg, Germany

A quantum dot attached to BCS superconducting leads exhibits a 0−π impurity quantum phase transition, which can be experimentally controlled either by the gate voltage or by the superconducting phase difference. From the theoretical point of view, the system is described by the single-impurity Anderson model, solving of which relies on heavy numerical methods. To enhance understanding and speed up characterization of the system, we aim to provide semi-analytical insight.

First, in the common case of the two leads having the same superconducting gap size, we explain the influence of asymmetric dot-lead coupling by relating the asymmetric system to a symmetric one. Surprisingly, it is the symmetric case, which is the most general, meaning that physical quantities in the case of asymmetric coupling are fully determined by their symmetric counterparts. We give ready-to-use conversion formulas for the 0−π phase transition boundary, on-dot quantities, and the Josephson current [1].

Next, we study the 0−π impurity quantum phase transition, clearly visible by a jump in the supercurrent in the ground state but smeared out to a crossover by finite temperature. For zero temperature, we present two simple analytical formulae describing the position of the phase boundary in parameter space for two opposing limits (the weakly correlated and Kondo regime). Furthermore, we show that the two-level approximation provides an excellent description of the low temperature physics near the phase transition. We discuss reliability and mutual agreement of available finite temperature numerical methods (the numerical renormalization group and quantum Monte Carlo) and suggest a novel approach for efficient determination of the quantum phase boundary from measured finite temperature data [2].

[1] A. Kadlecová, M. Žonda, T. Novotný, Phys. Rev. B 95(19), 195114 (2017).
[2] A. Kadlecová, M. Žonda, V. Pokorný, and T. Novotný, Phys. Rev. Applied 11, 044094 (2019).

Tuesday, July 2

Aspects of Topology and Quantum Interference in Single Molecule Transport

Daniel Hernangomez, **Ferdinand Evers**
University of Regensburg, Germany

Single molecules offer a fascinating testbed of effects that can be understood in terms of quantum interference and topology. The first part of the talk will recall counting rules that allow to predict occurrences of strong destructive interference in conjugate organic molecules. The second part is devoted to topological aspects of transport in quantum wires that can be rationalized within the Su-Schrieffer-Heeger model. An outlook towards experimental realizations will be offered at the end.

Self-consistent calculation of transport properties of molecular junctions

Vladislav Pokorný, Richard Korytár

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We calculate charge transport properties of molecular nano-junctions employing a combination of the density-functional theory (DFT) and a transport calculation based on the non-equilibrium Green functions technique, using our effective implementation of the non-equilibrium density matrix calculation. We focus on the effect of the spin-orbit coupling on the transport properties of short metallic nanowires and small organic molecules connected to large metallic leads. The thermodynamic limit of infinite leads is achieved using absorbing boundary conditions. The DFT solver and the transport code run inside a charge self-consistency loop, providing reliable results on the transmission function and magnetic properties.

Observation of electronic end-states in hydrogen-bonded organic 1D molecular chains on Au(111)

A. Cahlík (1), J. Hellerstedt (1), M. Švec (1), V.M. Santhini (1), S. I. Erlingsson (3), K. Výborný (1), P. Mutombo (1), J. Mendieta (1), S. Pascal (2), O. Siri (2), P. Jelínek (1)

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One-dimensional structures offer a rich ecosystem for realizing quantum states with potential application for advanced information technologies. Surface confined molecular self-assembly is one avenue for creating 1D systems, where the extant structure is controlled by the precursor shape, and functional group interlinking chemistry. As an example, recent studies of properly designed graphene nanoribbons demonstrated the presence of topologically protected end-states [1].

Here we study self-assembled 1D chains of zwitterionic molecule 2,5-diamino-1,4-benzoquinonediimines (DABQDI) [1] on Au(111) in ultrahigh vacuum at 5K using combined scanning tunneling and non-contact atomic force microscopies (STM/ nc-AFM) supported by theoretical analysis. Sub-molecular resolution achieved with a CO-functionalized tip hints structural information, specifically regarding the hydrogen bonds linking the precursor units. This is further supported by density functional theory (DFT) simulations and calculations of proton tunneling in between the unit cells, that enhances electronic communication across the chain.

On top of this, scanning tunneling spectroscopy (STS) measurements reveal a presence of in-gap electronic states near the Fermi energy, strongly localized to the chain ends.

To rationalize the presence of these states, we propose an extension of the seminal Su, Schrieffer, Heeger (SSH) [2] model by mimicking the chemical structure of 1D DABQDI chain. A detailed analysis of the proposed model shows possible solutions with non-trivial topological phase, that exist within intuitively reasonable range of model parameters.

[1] O. Groning et al, Nature 560, 209, 2018; D.J. Rizzo et al, Nature 560, 204 (2018).
[2] S. Pascal, O. Siri, Coordination Chemistry Reviews 350, 178-195, (2017).
[3] W. P. Su, J. R. Schrieffer, and A. J. Heeger, Phys. Rev. Lett. 42, 1698, (1979).

Isospin-flip spectroscopy

Nicolas Lorente

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E-20009 San Sebastian, Spain*

Degenerate orbitals in the presence of electronic correlation and an electron reservoir can behave like a two-level system. When the degeneracy is broken, this isospin character of the system is preserved albeit an energy cost. This gives rise to an inelastic effect with the peculiarity of a strong link between energy threshold and spatial distribution of the change in differential conductance. I will show the theory for an experimental system that displays this effect [PRL 121, 226402 (2018)].

Exchange rules for π-conjugated hydrocarbons

R. Ortiz (1,2,3), J. Fernández-Rossier (2)

(1) Department of Applied Physics University of Alicante, Spain, (2) QuantaLab International Iberian Nanotechnology Laboratory INL, Portugal, (3) Department of Physical Chemistry University of Alicante, Spain

A variety of planar π-conjugated hydrocarbons such as heptauthrene, Clar's goblet and, recently synthesized, triangulene have two electrons occupying two degenerate molecular orbitals. The resulting spin of the interacting ground state is often correctly anticipated as $S = 1$, extending the application of Hund's rules to these systems, but this is not correct in some instances. Here we provide a set of rules to correctly predict the existence of zero mode states, as well as the spin multiplicity of both the ground state and the low-lying excited states, together with their open- or closed-shell nature. This is accomplished using a combination of analytical arguments and configuration interaction calculations with a Hubbard model, both backed by quantum chemistry methods with a larger Gaussian basis set. Our results go beyond the well-established Lieb's theorem and Ovchinnikov's rule, as we address the multiplicity and the open-/closed-shell nature of both ground and excited states.

Following the Kondo temperature of a tunable single-molecule junction continuously from the Kondo singlet to the free spin 1/2 ground state

Martin Žonda (1,3), Oleksandr Stetsovych (2), Richard Korytár (3),
Markus Ternes (4,5), Ruslan Temirov (5,6), Andrea Racanelli (5,6),
F. Stefan Tautz (5,6), Pavel Jelínek (2,7), **Tomáš Novotný** (3), Martin Švec (2,7)

(1) Institute of Physics, Albert Ludwig University of Freiburg, Germany, (2) Institute of Physics, Czech Academy of Sciences, Prague, Czech Republic, (3) Department of Condensed Matter Physics, Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic, (4) Institute of Physics II B, RWTH Aachen University, Germany, (5) Peter Grünberg Institut, Jülich, Germany, (6) Jülich Aachen Research Alliance (JARA), Jülich, Germany, (7) RCPTM, Palacký University, Olomouc, Czech Republic

We monitor the conductance evolution by varying the coupling between an organic molecule and the Ag(111) surface by lifting the molecule from the surface with the tip of a scanning tunneling microscope. Numerical renormalization group theory analysis of the junction conductance reveals that the system is tuned from the strongly coupled Kondo singlet to the free spin 1/2 ground state. In the crossover between the strong and weak coupling regimes, where the Kondo temperature T_K is similar to the experimental T_{exp}, fitting procedures generically provide ambiguous estimates of T_K that tend to vary by an order of magnitude. Measuring the dependence of the junction conductance in external magnetic fields resolves this ambiguity if the Zeeman splitting due to the imposed magnetic field overcomes the thermal energy k_BT_{exp}. We demonstrate that Frota analysis overestimates T_K by several orders of magnitude in the transient and weak coupling regimes. The numerical analysis also allows us to determine molecular coupling to surface and tip and their evolution during the lifting process.

Spin correlations in graphene nanostructures probed with STM

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In this talk I will discuss the emergence of non-trivial quantum spin correlations in a variety nanographene nanostructures, using a full quantum description based on exact diagonalization of the Hubbard model, in a restricted Hilbert space. In addition, I will present our theoretical understanding of how STM spectroscopy can provide information about the spin excitations in these systems.

Non-covalent control of spin-state in metal-organic complex by positioning on N-doped graphene

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Nitrogen doping of graphene significantly affects its chemical properties, which is particularly important in molecular sensing and electrocatalysis applications. However, detailed insight into interaction between N-dopant and molecules at the atomic scale is currently lacking. Here we demonstrate control over the spin state of a single iron(II) phthalocyanine molecule by its positioning on N-doped graphene. The spin transition was driven by weak intermixing between orbitals with z-component of N-dopant (p_z of N-dopant) and molecule (d_{xz}, d_{yz}, $d_z{}^2$) with subsequent reordering of the Fe d-orbitals. The transition was accompanied by an electron density redistribution within the molecule, sensed by atomic force microscopy with CO-functionalized tip. This demonstrates the unique capability of the high-resolution imaging technique to discriminate between different spin states of single molecules. Moreover, we present a method for triggering spin state transitions and tuning the electronic properties of molecules through weak non-covalent interaction with suitably functionalized graphene.

Simulated STM fingerprints for transition-metal porphyrins and phthalocyanines on metallic surfaces

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Porphyrins and phthalocyanines with transition-metal cores make the ideal building blocks for functional thin films in nanotechnology. Although they were intensively studied in the past decades, some of their characteristics can only be described computationally. In this respect, we investigated the interaction characteristics between some transition-metal phthalocyanines/porphyrins and metallic surfaces i.e. Au(111) and Ag(111), with focus on simulated STM images to be further compared to experimental data. Our approach is based on the vdW-DF-cx exchange–correlation functional of Berland and Hyldgaard as implemented in SIESTA and includes DFT+U corrections. The simulation of the STM images was performed using the Tersoff-Hamman approximation as a "constant current" experiment. We emphasize the role of the transition metals (Cr, Mn, Fe, Co, Ni) as well as that of the organic moiety, and thus provide a powerful tool for discriminating between various experimental STM images.

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Interactions in superlattice quantum materials

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The conspiracy of electronic interactions, band structure, and lattice degrees of freedom forms a vital basis for exotic states of quantum matter. Here, we address the interplay of superlattice and interaction phenomena and discuss how electronic correlations and coupling between neighboring layers can be exploited to control quantum many-body states of layered van der Waals systems. First, we consider generic two-dimensional Mott-Hubbard systems and analyze possibilities of “Coulomb engineering”, i.e. controlling the electronic states realized in these systems via external screening. We then discuss the particular example of magic-angle twisted bilayer graphene (MA-tBLG), which has appeared as a rich ground for the realization of intricate superconducting, insulating and metallic many-electron states in a two-dimensional Dirac material framework. We analyze the interplay of internal screening and dielectric environment on the intrinsic electronic interaction profile of MA-tBLG and show that experimental tailoring of the dielectric environment presents a promising pursuit to provide further evidence for resolving the hidden nature of the quantum many-body states in MA-tBLG. Finally, we turn to emergent Mott-Hubbard states in charge density wave superlattices of group V TMDCs and discuss opportunities of realizing novel electronic quantum states, there.

To bend or not to bend? Correlation effects in low-dimensional silicon

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Despite the formal similarity, the structure, stability and chemical properties of unsaturated systems containing higher main group elements are strikingly different from their first-row counterparts. This is particularly evident in group IV, since the chemical flexibility of carbon is not shared by its congeners. In fact, the nature and structure of multiple bonds between higher main group elements remain elusive, thereby hindering the development of their low-dimensional systems. Here, focusing on the Si vs. C comparison, we shed light on this issue with the help of analytical modeling and first-principles calculations on both finite-sized and extended systems. We show that trans-bending in disilenes and buckling in low-dimensional silicon nanostructures (e.g., silicene) have a common origin, as both result from a tug of war between the σ and the π bonds in which the former favor distortion and the latter oppose to it. In carbon, π bonds are stiffer and effectively resist to the distortion, but in silicon they are softer and typically succumb. As a consequence, Si structures display weaker π bonds than they could otherwise do if undistorted. Correlation between π electrons plays a major role in this context, since Coulomb repulsion moves π bonds apart from the molecular orbital limit and renders them sensitive to doping charges. Hence, upon weakening the effective e-e repulsion in the p shell, one may completely remove any structural instability, strengthen the π bonds, and turn Si into a closer relative of C.

Electronic structure, optical and photocatalytic performance of SiC–MX_2 (M = Mo, W and X = S, Se) van der Waals heterostructures

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The stacking of monolayers in the form of van der Waals heterostructures is a useful strategy for band gap engineering and the control of dynamics of excitons for potential nano-electronic devices. We performed first-principles calculations to investigate the structural, electronic, optical and photocatalytic properties of the SiC–MX_2 (M = Mo, W and X = S, Se) van der Waals heterostructures. The stability of most favorable stacking is confirmed by calculating the binding energy and phonon spectrum. SiC–MoS_2 is found to be a direct band gap type-II semiconducting heterostructure.

Moderate in-plane tensile strain is used to achieve a direct band gap with type-II alignment in the SiC–WS_2, SiC–$MoSe_2$ and SiC–WSe_2 heterostructures. A difference in the ionization potential of the corresponding monolayers and interlayer charge transfer further confirmed the type-II band alignment in these heterostructures. Furthermore, the optical behaviour is investigated by calculation of the absorption spectra in terms of $\varepsilon_2(\omega)$ of the heterostructures and the corresponding monolayers. The photocatalytic response shows that the SiC–Mo(W)S_2 heterostructures can oxidize H_2O to O_2. An enhanced photocatalytic performance with respect to the parent monolayers makes the SiC–Mo(W)Se_2 heterostructures promising candidates for water splitting.

GeTe(111): direct evidence of static displacive ferroelectric response coupled to fully spin polarized bulk states

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Manipulating spin textures through electric fields is a timely issue due to the potential for applications in low power electronics. Recently, a novel class of ferroelectric materials combining ferroelectric order with Rashba type switching of spin textures at room temperatures [1-3] opened an intriguing route for a non-volatile static electrical control of the spin degrees of freedom. In this respect, GeTe(111) is a model system which appears to possess an extra bonus: fully spin-polarized bulk states. We extended the experimental verification of the ferroelectric switching of these fully spin-polarized states in spin and momentum-resolved ways [3] with operando x-ray photoelectron-diffraction experiments [4]. We found that the ferroelectric response feature systematic displacive response with characteristic fatigue effects consistent with operando spin-resolved measurements [3], proving that static electric fields indeed couple to GeTe domain polarizations and thus to Rashba-type splitting.

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Europium cyclooctatetraene nanowire carpets: a low-dimensional, organometallic ferromagnet

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Sandwich molecular wires are a particular 1D class of organometallic structures. They consist of a periodic sequence of 4f rare-earth metal cations, predominantly ionically bound and eight-fold coordinated to planar aromatic anions, based on the cyclooctatetraene (C_8H_8 (Cot)) molecule as a ligand. Because of organometallic hybridization between the metal atomic states and the extended π orbitals of the Cot, the metal ions in the wire were proposed to couple magnetically [1]. Here we investigate the magnetic and electronic properties of europium cyclooctatetraene (EuCot) nanowires by means of low-temperature X-ray magnetic circular dichroism (XMCD) and scanning tunneling microscopy (STM) and spectroscopy (STS) [2]. The EuCot nanowires are prepared in situ on a graphene surface. STS measurements identify EuCot as an insulator with a minority band gap of 2.3 eV. By means of Eu $M_{5,4}$ edge XMCD, orbital and spin magnetic moments of (-0.1 ± 0.3) μ_B and $(+7.0\pm0.6)$ μ_B, respectively, were determined. Field-dependent measurements of the XMCD signal at the Eu M_5 edge show hysteresis for grazing X-ray incidence at 5 K, thus confirming EuCot as a ferromagnetic material. Our density functional theory calculations reproduce the experimentally observed minority band gap. Modeling the experimental results theoretically, we find that the effective interatomic exchange interaction between Eu atoms is on the order of millielectronvolts, that magnetocrystalline anisotropy energy is roughly half as big, and that dipolar energy is approximately ten times lower. We are confident that the finding of ferromagnetic ordering in an experimentally well-accessible, surface-supported, organometallic system will provide new inspiration for the field of molecular spintronics.

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Tuning electronic transport of 1D coordination polymers by the choice of the transition metal: Fe, Co and Ni

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The choice of the transition metal atom in an organometallic complex decisively affects its physiochemical properties such as its color or magnetic moment. Towards rationalization of the impact of the choice of transition metal atom on the electronic and spintronic properties of organometallic wires, we study the electric transport through Fe, Co and Ni containing 1D coordination polymers contacted by the tip of a scanning probe microscope (SPM). The coordination polymers are synthesized in-situ by co-deposition of the metal atoms and the quinonediimine (2,5-diamino-1,4-benzoquinone-diimine) ligand onto Au(111). Scanning tunneling microscopy combined with atomic force microscopy is used for characterization as well as lifting and transport experiments. All three coordination polymers are found to be structurally equivalent, with a coordination geometry consistent with the expected formal M(II) oxidation states. This implies dehydrogenation of the quinonediimine ligand. Wires with lengths over 100 nm can be obtained. Transport measurements are performed by contacting one end of the surface-adsorbed polymer with the SPM tip. The combination of STM and AFM allows for simultaneous measurement of the current, conductance and force gradient as a function of bias voltage and lifting height. The transition metal element contained in the organometallic wire is found to have a profound impact on its electronic properties: lifting of Fe and Ni wires leads to the opening of a band gap at heights of a few nanometers. In contrast, Co containing polymers do not exhibit no gap. In addition, conductance measurements reveal pronounced steps at bias voltages of a few millivolts for Fe and Co wires but not for Ni ones. These conductance steps are tentatively attributed to spin excitations.

Element-Specific Magnetism of Thulium-Cyclooctatetraene Nanowires

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The analysis of the magnetic coupling and the magnetic anisotropy of localized 4f magnetic moments connected via organic ligands in molecular networks is a highly topical field of research owing to potential applications in organic spintronics. Our on-surface synthesis method allows us to synthesize sandwich-molecular nanowires that consist of alternating ring-like cyclooctatetraene (Cot) molecules and varying rare-earth ions. Using thulium hereby offers not only various phases but may also exhibit interesting properties due to its strong single-ion anisotropy compared to e. g. europium. The electronic and magnetic properties of these compounds were studied by X-ray absorption spectroscopy and X-ray magnetic circular dichroism accompanied by a theoretical approach based on multiplet calculations. The X-ray absorption spectroscopy of the systems indicate a $4f^{12}$ electronic state of the thulium. In contrast to Eu-cyclooctatetraene nanowires, the XMCD of the Tm systems suggest, that they are not saturated even in high fields and low temperatures. We acknowledge financial support by DFG through the project WE 2623/17-1.

Ab-initio treatment of the Lanthanides: from solids to clusters and atoms

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Ab-initio calculations based on density-functional theory have become a fundamental tool to obtain the quantum mechanical properties of a variety of systems without adjustable parameters. Unfortunately, systems containing partially-filled shells of localized electrons, such as 3d or 4f, have been difficult to investigate, due to the problems related to the treatment of strong electronic correlations. Nevertheless, in the last decade many methods were proposed to overcome these limitations, and overall much progress was made. In this presentation, I will show how a combination of density functional theory and dynamical mean-field theory can be used to investigate rare-earth based systems. I will illustrate how this approach can provide an invaluable insight into electronic, magnetic and structural properties of solids, clusters and adatoms. I will explain how one can combine this technique with advanced methods for spectroscopy and magnetization dynamics. I will finally demonstrate how the aforementioned calculations can be exploited for materials discovery and engineering via 'big data' analysis.

Influences of vacancy and doping on electronic and magnetic properties of monolayer SnS

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Based on the first-principles calculations, we investigate the structural, electronic, and magnetic properties of defects in monolayer SnS. We study the formation and migration of vacancies at both Sn- and S-sites. In comparison to the S-site vacancy, our calculations show that creating a vacancy at the Sn-site requires lesser energy, indicating that the vacancy at the Sn-site is more likely to be formed in experiments with energetic particle irradiation. For the Sn-rich (S-rich) environment, the vacancy at the S-site (Sn-site) is more likely to be found than the vacancy at the Sn-site (S-site). Reducing the formation of vacancy clusters, the S vacancy remains at the position where it is created because of the high vacancy migration barrier. Both types of vacancies remain nonmagnetic. To induce magnetism in monolayer SnS, we also study the transition metal (TM=Mn, Fe, and Co) doping at the Sn-site and find a significant influence on the electronic and magnetic properties of monolayer SnS. The doping of TM changes non-magnetic monolayer SnS to magnetic one and keeps it semiconducting. Additionally, long-range ferromagnetic behavior is observed for all the doped system. Hence, doping TM atoms in monolayer SnS could be promising to realize a two-dimensional diluted magnetic semiconductor. More interestingly, all the doped TM configurations show a high spin state, which can be used in nanoscale spintronic applications such as spin-filtering devices.

Ge-vacancy complexes: a viable route toward quantum information processes at room temperature

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The development of on-demand individual deep impurities in silicon is motivated by their employment as a physical substrate for qubits [1], nanoscale transistors [2], single photon emitters [3] and Hubbard simulators [4]. Single-atom silicon devices based on conventional doping elements such as phosphorous [5-7] can operate only at cryogenic temperature due to shallow weakly localized ground state impurity levels.

Differently, the implantation of Ge dopants in silicon and the subsequent annealing generate stable Ge-vacancy defects [8] that are promising candidates to achieve single-atom quantum effects at room temperature. These hybrid complexes combine indeed the properties of the silicon vacancy, which carries deep states in the band-gap, with the accurate spatial controllability of the defect obtainable through state of the art single-ion implantation of Ge atom.

We characterize GeV_n complexes in silicon by means of accurate ab initio density functional theory calculations with a hybrid functional. We get insight on the defect local arrangement and on their electronic properties, demonstrating their suitability for the purposed application. We show that due to the deep defect levels the electrons bound to the defects are strongly correlated and more localized than in conventional dopants [9]. Through a multi-scale theoretical approach that combines ab initio DFT and an extended Hubbard model we analyze also the electronic transport through a chain of GeV dopants. By mapping the ab initio results into the model Hamiltonian we are able to describe the electronic conductance of the array as a function of the temperature and to determine the activation energy for hopping processes.

We compare the theoretical results with the experimental data showing a notable agreement between the calculated and measured excited state energies and temperature activated transport properties [10].

This work has been performed in the context of a user access request within the Nanoscale Facilities and Fine Analysis project [11], funded by European Community.

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Direct Imaging of Orbitals using Inelastic X-Ray Scattering

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It is generally accepted that the correlated motion of electrons is at the origin of many interesting physical properties including unconventional superconductivity, colossal magnetoresistance, and multiferroicity, just to name a few. These spectacular properties emerge through intricate interplay of charge-spin-orbital degrees of freedom of valence electrons, so their characterization is an essential ingredient for modeling these curious behaviors and revealing the underlying mechanisms. This is a very challenging and delicate undertaking, which often requires the knowledge of the active valence wave function.

We developed a new experimental method that directly images the active orbital in solids, without advanced calculation or spectroscopic analysis [1]. The method, s-core-level non-resonant inelastic X-ray scattering (s-NIXS), relies on high momentum transfer in the inelastic scattering process, which is necessary for dipole-forbidden terms to gain spectral weight. To demonstrate the strength of the technique, we imaged the text-book example, x^2-y^2/$3x^2$-r^2hole orbital of the Ni^{2+} ion in NiO single crystal. We will present the basic principles of s-NIXS and its experimental implementation. We will also show how we can apply this technique to unveil the active orbitals in complex oxides as well as to determine the orbital character in highly metallic systems such as elemental Cr, Fe, and Ni.

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Impact of the magnetic dipole term T_z on x-ray magnetic circular dichroism spectra of low-dimensional systems

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Magnetism of diluted low-dimensional systems and nanostructures is often studied by means of X-ray magnetic circular dichroism (XMCD). The analysis of XMCD experiments is greatly aided by the XMCD sum rules which, in principle, give access to the spin and orbital magnetic moments of the chemically specific photoabsorbing atom. Unfortunately, the XMCD spin sum rule allows for the spin magnetic moment m_{spin} to be determined only in combination with the magnetic dipole term T_z. This T_z term is not just a minor factor that affects the sum rules analysis; on the contrary, it may change the overall picture completely. We present few examples demonstrating that neglecting the T_z term could in some cases lead to completely wrong conclusions about the trends of m_{spin} with the size of the system or with the direction of the magnetization.

Earlier it was shown that the magnetic dipole term T_z can be eliminated from the sum rule analysis altogether by employing a special experimental setup. We demonstrate that for supported nanostructures this can be done only if certain conditions concerning the strength of the spin-orbit coupling and the hybridization between the states of the nanostructure and of the substrate are satisfied.

Shift of Core-Electron Energy Levels by Next Nearest Neighbors

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By means of density functional theory calculations, we explain the anomalous thermally induced peaks in the XPS spectra of the core electrons previously reported for a metalloporphyrin molecule adsorbed on TiO_2 surface [JCP 146, 184704 (2017)]. The shift of such levels is routinely interpreted as a signature of charge exchange with the substrate. However, in this case we show that some atoms with no chemical bonding to the surface also experience significant core level shifts. Our interpretation based by simulations is that the electronic structure of this dye synthesizer-semiconductor complex is partly originated from its core geometry deformation. In particular, the next neighbors are also affected. One should therefore use the valuable photoelectron spectroscopy outputs with caring such effects. Our theoretical prediction is in fair agreement with experiment, based on which we are able to disclose the mechanism of the splitting the degenerate core states in terms of the symmetry breaking rather than the dye-substrate chemical bonding. This, additionally, shows that how one can facilitate the high sensitivity of photoelectron spectroscopy to help identification the adsorption site and orientation of molecules on atomic surfaces.

LDA+DMFT approach to resonant inelastic X-ray scattering in correlated materials

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We develop an ab-initio computational method for analyzing modern resonant inelastic X-ray scattering (RIXS) spectra in correlated materials, such as 3d transition-metal and

4f rare-earth compounds. The method builds on the combination of local density approximation (LDA) and dynamical mean-field theory (DMFT), and can describe fine RIXS features originating from both localized (e.g. multiplet excitations) and delocalized excitations (e.g. charge-transfer, unbound electron-hole pair excitations) [1]. In this talk, we present calculated L-edge RIXS spectra for typical 3d transition-metal oxides: NiO, Fe_2O_3, $LaMnO_3$ and $RNiO_3$ with a comparison to high-resolution experimental data. We discuss the relation of the emergent charge excitations in RIXS spectra and low-energy valence states. Special attention will be paid on the coexistence/interference between the localized and delocalized excitations in the RIXS spectra, observed in recent experiments [2].

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[2] V. Bisogni et al. Nat. Commun. 7. 13017 (2016).

The theory of continuum charge excitations in high-valence transition-metal oxides revealed by resonant inelastic x-ray scattering

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We present a computational study of L-edge resonant inelastic x-ray scattering (RIXS) in 3d high-valence transition-metal (TM) oxides: $RNiO_3$ (R: rare-earth element); $LaCuO_3$ and $NaCuO_2$ [1]. We employ a theoretical approach based on local density approximation (LDA) and dynamical mean-field theory (DMFT), where the Anderson impurity model with DMFT hybridization function is extended to include TM 2p core orbitals. The heart of the present approach is the configuration-interaction-based exact-diagonalization impurity solver that allows to include the low-energy electron-hole pairs accurately, necessary to describe RIXS in the high-valence materials with negative charge-transfer energy. The LDA+DMFT approach enables to describe both bound and unbound electron-hole pair excitations in the RIXS spectra. In this talk, we discuss the behavior of the fluorescence-like (FL) feature and how it is connected to the low-energy valence states and crystal structure. Special attention will be paid to the selection rules for the visibility of the FL RIXS feature, that reflect the lattice geometry around the x-ray excited TM site. Our results reproduce the recent experimental data for $RNiO_3$ [2].

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Impurity scattering and Friedel oscillations in model correlated fermionic systems

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Scattering from the impurities and the resulting Friedel Oscillations (FO) in the Fermi liquid phase, Mott insulating phase, and at the Mott transition is studied in finite lattice systems using the one-band Hubbard model, both at zero and finite temperatures in the presence of a single impurity potential [1,2,3]. The problem is also extended for the cases of multiple impurities. These impurities model the effects of defects and adsorbed atoms on the surfaces of real systems with correlated electrons. Electronic correlations are accounted for by solving the real-space dynamical mean-field theory (R-DMFT) equations, and also using other reliable yet computationally cheap self-energy approximations based on the R-DMFT. It is seen that the FO is damped with the interaction and the Fermi liquid renormalization factor is primarily responsible for this damping. FO almost disappears at the Mott transition and completely beyond it in the Mott insulating phase. The screening charge decreases with the interactions and approaches zero towards the Mott transition. It is left as an open question if any signatures of FO can be observed in real Mott systems, e.g. transition metal oxides which are the potential functional materials for the Mott transistors [4,5]. Moreover, it is interesting to probe if these quantum oscillations would affect the different transport properties, e.g. transport current, resistivity etc. in such systems.

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Thursday, July 4

Actuating and probing a single-molecule switch at femtosecond timescales

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Accessing ultra-fast non-equilibrium phenomena is enabled by terahertz (THz) scanning tunneling microscopy [1] (THz-STM) through combining STM with lightwave electronics. In THz-STM, the electric field of a phase-stable single-cycle

THz waveform acts as a transient bias voltage across an STM junction. In analogy to the all-electronic pump-probe scheme introduced recently in STM [2] these voltage transients may result in a net current that can be detected by time-integrating electronics.

The recent development of this lightwave STM has enabled the combined femtosecond and sub-angstrom resolution in observing matter [3]. We now demonstrate the first combined femtosecond and sub-angstrom access in the control of matter. Ultrafast localized electric fields in lightwave STM enable exerting atom-scale femtosecond forces to selected atoms. By shaping atomic forces on the intrinsic timescale of molecules, coherent atomic motion can now be excited. Utilizing this coherent structural dynamics, we can modulate the quantum transitions of a single-molecule switch by up to 39%. We directly visualize the coherent excitation of the switch in the first femtosecond single-molecule movie [4].

To resolve the impact of coherent control of the single-molecule switch, alongside, we introduce single-shot action spectroscopy in lightwave STM as the first concept resolving individual path-selective reaction events of a single molecule in space and time. With this novel concept, we detect the outcome of every single laser shot and further separate the statistics of the two inverse reaction paths. Our results open a new chapter in the control and observation of reactions of individual molecules directly on the relevant ultrafast and ultrasmall scales.

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Transmission Spectroscopy of Molecular Spin Ensembles through Microwave Planar Resonators

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Over the last decade molecular spins have been object of intense research as potential candidates for quantum technologies [Ghirri et al., Magnetochem. 3, 12 (2017)]. The interest is given by the possibility of tailoring their magnetic properties at synthetic level as well as their relatively easy integration on surfaces. Another key aspect is the presence of both electronic and nuclear spin degrees of freedom which, in principle, might open wide possibilities for the implementation of protocols for quantum information processing. Here, two of the main challenges are addressing the transitions with external microwave and radiofrequency stimuli and developing suitable protocols for the manipulation of the spins.

In this work, we deal with the microwave excitation of molecular spin ensembles at low temperatures. To achieve this goal, we have first developed a set of planar superconducting resonators at microwave frequency to couple to the ensembles via magnetic dipolar interaction in an electron spin resonance-like transmission spectroscopy [Bonizzoni et al., Adv. in Phys. X 3, 1435305 (2018)]. The coupling conditions of several molecular ensembles are first studied under the continuous-wave excitation regime. Remarkably, experimental evidence of coherent spin-photon coupling between the ensembles and the resonators at low temperatures is found within two different approaches. In the first one the number of coupled spins of a diluted Vanadyl Phthalocyanine is maximized by using an optimized resonator [Bonizzoni et al., Sci. Rep. 7, 13096 (2017)], while in the second one the exchange narrowing effect of the spin linewidth in concentrated organic radicals is exploited [Ghirri et al., Phys. Rev. A 93, 063855 (2016)]. We then focus to experiments in the pulsed-wave regime. Here an Arbitrary Waveform Generator is used for the synthesis of the microwave pulses which are then injected into the resonant geometry to drive the evolution of the spins. Our preliminary results concerning the measure of the phase memory time on an ensemble of Vanadyl Phthalocyanines are then given.

Spin, angle and time-resolved photoemission studies of WTe_2

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Molybdenum dichalcogenides are probably the most studied single layer TMDCs by virtue of being appealing for sundry possible applications suchlike transistors, diodes, solar cells or more fundamental studies of spin or valley pseudospin and their interactions. Tungsten-based counterparts are on the other hand evincing much stronger spin-orbit coupling due to which all the spin-related effects are more stable at room temperature and thus more feasible for application. WTe_2, the type-II Weyl semimetal is in particular interesting due to having two pairs of spin-differentiated Weyl points above Fermi energy. We have conducted several experiments following the evolution of the band dispersion in the vicinity of X and Y points of the Brillouin zone of WTe_2 which is substantial for understanding the fundamental properties of the structure-property relation of the system. Ab-initio set of photoemission calculations was performed using SPR-KKR package and compared to photoemission experimental results. In adition, TR-SARPES studies were carried out by the group on such materials to determine the ultrafast dynamics of the carrier density and to disentangle the subsequent relaxation processes using pump-probe experiments.

Towards predictive theory of two-particle spectroscopies in strongly correlated materials

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Two-particle (2P) excitations provide the prime path to probe and to manipulate electronic states of materials. Examples include optical experiments, electron loss spectroscopy, inelastic x-ray or neutron scattering. Theoretical description of 2P excitations in correlated materials is notoriously difficult as these involve unbound electron-hole pairs as well as bound pairs such as spin waves or excitons. The behavior of 2P excitations is particularly interesting in states with spontaneously broken symmetry where gapless collective modes are predicted by symmetry considerations. I will present dynamical mean-field calculation of collective modes in excitonic magnets at intermediate and weak coupling to demonstrate capability of this approach. As a material specific example, I will present simulation of RIXS spectra of $LaCoO_3$ where spinful excitons where recently observed.

Electronic and spectral properties of clean and C_{60}-covered atom-thick Chromium oxide at the Fe(001) surface

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Chemisorption of a single atomic layer of oxygen on the Fe(001) surface yields a highly ordered and reproducible benchmark substrate [1] for theoretical and experimental studies, and for the epitaxial growth of metal oxides, including atom-thick Cr_xO_y layers, and hybrid interfaces with foreseen applications e.g. in organic spintronics.

This talk initially presents ab initio investigations that have supplemented microscopy and spectroscopy experiments of the electronic and magnetic properties of two-dimensional Chromium oxides of Cr_3O_4 and Cr_4O_5 stoichiometry grown on Fe(001), featuring antiferromagnetic magnetic configurations with underlying Fe(001) [2,3]. Despite Cr / CrO systems are notoriously difficult for mean field approaches, generalized-gradient results are found to explain most experimental findings, with a rigid shift of oxygen bands accounting for electronic correlation effects.

We eventually consider the effect of inserted Cr_4O_5 layers at the interface between the prototypical C_{60} organic semiconductor and Fe(001), which is shown to enhance the magnetic hybridization between the molecule and the surface through X-ray Magnetic Circular Dichroism (XMCD) [4,5]. By means of ab initio calculation we characterize the local interface morphology, the magnetic configuration of the surface and the induced spin dependent electronic properties of the molecule, the latter reflecting the magnetic electronic properties of the surface at the relevant energy range. As seen from the substrate, adsorbates can influence the magnitude and even orientation of surface Cr magnetic moments. The interest in this interface is then twofold: on one side the thin magnetic oxide allows tailoring the magnetic properties of the organic layer, on the other side the adsorption of C_{60} can be envisioned as a tool to control the magnetic ordering of Cr atoms at the interface.

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Magnetic anisotropy of UO_2 studied on UO_2/Fe_3O_4 thin films

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Large magnetic anisotropy (MA) of the material is a keystone to the high-density magnetic storage. It defines the easy magnetization direction and ensures information storage by preventing the magnetization from switching. In bilayers, MA can be controlled via pinning of a ferromagnetic (F) layer by an antiferromagnet (AF) in the exchange bias (EB) [1] systems [2]. In this work we aim at evaluating the anisotropy constant in the antiferromagnetic uranium dioxide by making use of exchange bias effect study in the UO_2/Fe_3O_4 bilayers deposited on various substrates. The shift of the magnetic hysteresis curve along the field direction of the magnetic bilayer field-cooled below the Néel temperature (T_N) of the antiferromagnet is related to its anisotropy constant K_{AF} [3,4] via the critical thickness of the AF layer, above which the antiferromagnet is strong enough to pin the magnetization of a ferromagnet. Using reactive sputtering from metallic U and Fe targets, we prepared sets of the UO_2/Fe_3O_4 samples with the varied thickness of UO_2 (50–300 Å) and with a constant thickness of magnetite (~270±20 Å). The stoichiometry of each deposited layer was controlled by X-ray photoelectron spectroscopy (XPS). The samples were further characterized by Rutherford back-scattering spectrometry (RBS) and x-ray diffraction (XRD). Magnetization study revealed a large EB in the samples [5]. Using the approach of Ref. 4, we find the anisotropy constants of UO_2 in the samples ~0–1-0.3 MJ/m^3. We further compare the experimental results with those obtained theoretically using a first-principles density-functional + dynamical-mean-field theory (DFT+DMFT) method [6] in conjunction with a quasi-atomic (Hubbard-I) treatment of U 5f states. We predict a strong easy-axis anisotropy to originate from the substrate-induced in-plain tetragonal compression of the UO_2 layer.

The samples were prepared in the framework of the TALISMAN project of the European Commission Joint Research Centre, ITU Karlsruhe. RBS measurements have been carried out at the CANAM (Centre of Accelerators and Nuclear Analytical Methods) infrastructure LM 2015056 and supported by OP RDE, MEYS, Czech Republic under the project CANAM OP, CZ.02.1.01/0.0/0.0/16_013/0001812 and by the Czech Science Foundation (GACR 18-03346S and 18-02344S). Part of the work was supported by “Nano-materials Centre for Advanced Applications,” Project CZ.02.1.01/0.0/0.0/15_003/0000485, financed by ERDF.

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Thermal displacements in microscopies and spectroscopies

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Thermal mean square displacements have the wonderful musty smell of the 1910s, when Debye showed a simple classical harmonic model could explain the form factor for the scattering of X-rays (or neutrons or anything small) from a crystalline lattice. These effects have had a resurgence in the past few years, as the precision and power of ab initio methods has grown, and meeting experiment now requires taking realistic environmental constraints into account. Vibrational modes are routinely available for complex materials, through density functional perturbation theory, and even beyond with anharmonic fitting methods.

I will show examples of the consequences of thermal vibrations on the electronic and magnetic properties of bulk and nanostructured materials. The interaction of vibration waves with magnons is an important ingredient in modern spin(calori)tronics devices: we estimate thermal effects on magnons in a renormalized atomistic spin dynamics framework, and show its strong interference with disorder in bulk permalloy [1]. In nanostructured materials, thermal displacements can become more strongly anisotropic, with important effects both in microscopy and spectroscopy. We quantify their impact on the movement of defects in graphene in a TEM [2], and the X-ray photoemission spectrum of noble metal surfaces [3].

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Phonon-induced electronic relaxation in a strongly correlated system: the Sn/Si(111) (√3 x √3) adlayer revisited

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The ordered adsorbate layer Sn/Si(111) (√3 x √3) with coverage of one third of a monolayer is considered as a realization of strong electronic correlation in surface physics. Our theoretical analysis shows that electron-hole pair excitations in this system can be long-lived, up to several hundred nanoseconds, since the decay into surface phonons is found to be a highly non-linear process.

We combine first-principles calculations with help of a hybrid functional (HSE06) with modeling by a Mott-Hubbard Hamiltonian coupled to phononic degrees of freedom. The calculations show that the Sn/Si(111) (√3 x √3) surface is insulating and the two

Sn-derived bands inside the substrate band gap can be described as the lower and upper Hubbard band in a Mott-Hubbard model with U=0.75eV. Furthermore, phonon spectra are calculated with particular emphasis on the Sn-related surface phonon modes. The calculations demonstrate that the adequate treatment of electronic correlations leads to a stiffening of the wagging mode of neighboring Sn atoms; thus, we predict that the onset of electronic correlations at low temperature should be observable in the phonon spectrum, too. The deformation potential for electron-phonon coupling is calculated for selected vibrational modes and the decay rate of an electron-hole excitation into multiple phonons is estimated, substantiating the very long lifetime of these excitations.

Theoretical Study of Structural, Electronic, Vibrational, Mechanical and Thermoelectric Properties with high mobility carrier of ternary Compounds

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Using the first-principle methods and deformation theory, we detail investigate the two-dimensional semiconductor materials with monolayer compounds which suggest a novel playground to implement nanoscale mechanical, thermoelectric and electronics devices to improve their functionality. Using the combination of approaches to compute the electronic and phonon structures with Green's function-based transport techniques, we report the thermometric performance of the group-III ternary monochalcogenides compounds. Our outcomes show strong mechanical properties and high carrier mobility (~ $2.18\text{x}10^5$ $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$). Our materials also show the high figure of merit at room temperatures ZT ~1.12. Thus, indicating at the high potential of these new materials in thermoelectric application.

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More surprises in f-electron magnetism

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Surface supported single magnetic atoms, the so-called "single-atom magnets", open new opportunities in a quest for the ultimate size limit of magnetic information storage. Initially, the research mainly focused on 3d-atoms on surfaces. Recently, the attention was turned to the 4f-atoms, culminating in the experimental discovery of magnetically stable Ho atom on MgO(001) substrate, and Dy atom on graphene/Ir(111).

I address the electronic and magnetic character of 4f-atoms on metal and Graphene substrate making use of a combination of the DFT+U with the exact diagonalization of Anderson impurity model (DFT+U+ED). The spin and orbital magnetic moments of Dy@Ir(111) and Dy/graphene/Ir(111) are evaluated and compared with experimental XMCD data. The magnetic anisotropy energy is estimated, and the magnetic stability is discussed. The role of 5d-4f interorbital exchange polarization in modification of the 4f-shell energy spectrum is emphasized.

Implementation of DFT+Hubbard-I in the FLAPW-method and application to 4f adatoms

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The goal of my master's thesis is to implement the LDA+Hubbard-I method in the framework of the FLAPW code FLEUR, which is developed at the IAS-1 Institute at the FZ Jülich. For this purpose, it is also necessary to implement functionality for calculating green's functions in the FLAPW basis. In the following, I want to apply this method to systems like Gd on Cu.

FLEUR is a full potential linearized augmented plane wave code which is applicable to a wide range of structures and elements. The current development of the code is aimed at high-throughput computations [1].

The LDA+Hubbard-I method works in the atomic limit of DMFT and is specifically developed for 4f systems, where we can neglect the hybridization effects of the 4f orbitals [2]. The advantage we gain from this method is that it is computationally relatively cheap when compared to other DMFT methods while including dynamical effects from the correlated electrons. The latter are ignored in the LDA+U method.

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Controlling fundamental electronic interactions in $SrTiO_3$ thin films by Ni doping

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Strontium titanate ($SrTiO_3$) has been a subject of intensive discussion in recent years both experimentally as well as theoretically. Strontium titanate is an insulator with a large band gap ($E_g = 3.2$ eV), but it can become conductive by doping with transition metals or oxygen vacancies [1]. Here we report the fabrication and electronic properties of Ni doped $SrTiO_3$ polycrystalline and crystalline films. The polycrystalline films were prepared by reactive magnetron co-sputtering with different Ni concentration. Whereas the crystalline films were grown by pulsed laser deposition (PLD). In this experimental investigation, we used high energy ARPES and core level spectroscopy to perform the electronic properties of the films as a function of doping concentration. Our experimental results were supported by ab-initio calculations using SPR-KKR's one-step model of photoemission [2,3].

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