Sino-German Symposium on

Computational Materials Science: Methods and Applications

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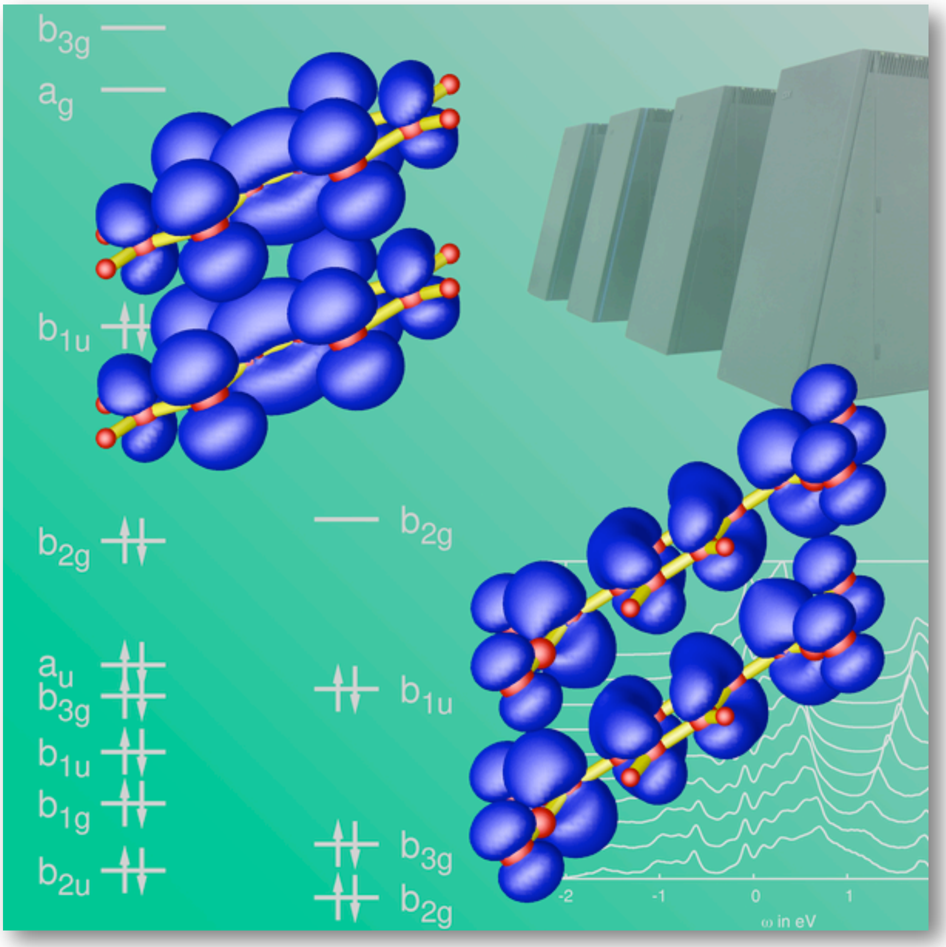

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Program

Monday, 21 September		
8:30	Welcome	
chair:	E. Koch	
9:00	V. Blum	Density functional theory and beyond with FHI-aims: Concepts and applications
9:40	L.X. He	Method to construct transferable minimal basis sets for ab initio calculations
10:20	J.L. Yang	Nearly free electron states in gated graphene nanoribbon superlattices
11:00	Coffee	
11:20	O. Gunnarsson	Electron-phonon coupling in strongly correlated systems
12:00	Z. Fang	LDA+Gutzwiller method for correlated electron systems
12:40	Lunch	
chair:	S.H. Wei	
14:30	X.G. Wan	Calculated magnetic exchange interactions in high-temperature superconductors
15:10	E. Pavarini	Origin of orbital-order in $KCuF_3$ and $LaMnO_3$
15:50	Coffee	
16:10	Z.Y. Lu	Electronic structure and magnetic properties of iron pnictides
16:50	O.K. Andersen	Explaining the band structure and magnetism of the new iron superconductors
17:30	H.O. Jeschke	Ab initio molecular dynamics for structural properties of layered Mott insulators and iron pnictides
18:10	Discussions & Dinner	

Tuesday, 22 September		
chair:	R.O. Jones	
9:00	Q. Sun	Computational design of materials for hydrogen storage in quasi-molecular form
9:40	S. Blügel	Computational design of materials for spintronics
10:20	W.Q. Zhang	Search for new thermoelectric materials based on electronic structure calculations
11:00	Coffee	
11:20	Z. Zeng	Characterizing graphene by doping
12:00	D.Y. Sun	Capillary force-induced structural instability in liquid encapsulated elastic circular tubes
12:40	Lunch	
chair:	R.Q. Wu	
14:30	A. Dolfen	Realistic description of strongly correlated organics
15:10	S. Chen	First-principles study of multiternary semiconductors and their alloys
15:50	Coffee	
16:10	R.O. Jones	Molecular dynamics/density functional (MD/DF) calculations for phase change memory materials: progress and promise
16:50	S.H. Wei	Computational study of unusual material properties of oxides
17:30	Dinner	

Wednesday, 23 September		
chair:	Z. Fang	
9:00	J. Ni	Boron and boron-carbon nanostructures
9:40	Z.Q. Yang	First principles investigation on quantum transport of atomic and molecular wires
10:20	S. Andergassen	Correlation effects on transport through quantum dots and wires
11:00	Coffee	
11:20	E. Koch	Quantum cluster extensions of dynamical mean-field theory
12:00	E. Gorelov	Continuous-time quantum Monte Carlo solvers for DMFT
12:40	Lunch	
chair:	J.M. Dong	
14:30	S. Heinze	Understanding spin-sensitive scanning probe microscopy based on electronic structure theory
15:10	R.Q. Wu	Determination of structural and magnetic properties of dilute magnetic semiconductors with x-ray magnetic circular dichroism
15:50	Coffee	
16:10	D. Zhang	Finite element method for solving Kohn-Sham equations based on self-adaptive tetrahedral mesh
16:50	R. Zeller	Methodology and capabilities of the KKR Green function method
17:30	W.N. E	Recent progress on linear scaling methods for electronic structure analysis
18:10	Discussions & Dinner	

Thursday, 24 September		
chair:	O.K. Andersen	
9:00	G. Bihlmayer	Unique magnetic spiral structures in low dimensions
9:40	J.M. Dong	Spin-orbit coupling in carbon-based nanostructured materials
10:20	Z.P. Liu	Electrocatalysis: going to atomic level
11:00	Coffee	
11:20	D.J. Shu	First principle calculation of the vacancy formation on TiO_2 surfaces
12:00	X.G. Gong	Predictive Modeling of Novel Structures and Structural Phase Transitions in Zero and One Dimensions
12:40	Lunch	
14:30	visit Fudan University	
	Banquet, Yu-Yuan Garden	
	boat trip through The Bund and Pudong	

Abstracts

Correlation effects on transport through quantum dots and wires

Sabine Andergassen
Institut für Theoretische Physik A, RWTH Aachen

The low-energy transport properties of mesoscopic systems are strongly influenced by electron correlations. The effects arising on different energy scales can be described very efficiently by renormalization-group techniques. We will focus on the functional renormalization group as a promising approach to investigate more complex geometries, and present applications to quantum dots and wires illustrating the interplay of Kondo physics and Luttinger-liquid behavior.

Explaining the band structure and magnetism of the new iron superconductors

Ole Krogh-Andersen
Max-Planck-Institut für Festkörperforschung, Stuttgart

Unique magnetic spiral structures in low dimensions

Gustav Bihlmayer
Institute for Advanced Simulation and Institut für Festkörperforschung, Forschungszentrum Jülich

Magnetic nanostructures are often complex, multicomposite materials with low symmetry, non-collinearity and strong spin-orbit coupling effects. In the last years we could establish first-principles methods to describe accurately the underlying magnetic interactions, i.e. the symmetric and antisymmetric exchange, the magnetocrystalline anisotropy and their dependence on dimensionality and structure in realistic materials. This talk will focus on low-dimensional magnetic systems and the importance of relativistic effects for the magnetic properties: due to the lack of inversion symmetry at the surface, the Dzyaloshinskii-Moriya interaction can induce non-collinear spin structures of a particular rotational sense in ultrathin magnetic films. Different computational aspects within the context of the full-potential linear augmented plane-wave (FLAPW) method will be presented [1]. As applications three examples will be discussed, domain walls in two monolayers Fe on W(110) [2], an antiferromagnetic monolayer Mn of W(110) [3], and the ferromagnetic Mn on W(100) [4].

[1] M. Heide, G. Bihlmayer, and S. Blügel, Physica B **404**, 2678 (2009)
[2] M. Heide, G. Bihlmayer, and S. Blügel, Phys. Rev. B **78** 140403(R) (2008)
[3] M. Bode, M. Heide, K. v. Bergmann et al., Nature **447**, 190 (2007)
[4] P. Ferriani, K. v. Bergmann, et al., Phys. Rev. Lett. **101**, 027201 (2008)

Computational Design of Materials for Spintronics

Stefan Blügel
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Magnetic storage and magnetic recording is world wide by far the most important way to save data and information over long times. The quest to store information in increasingly smaller volumes, to read, write and process information faster and with decreasing usage of energy in combination with the non-volatile properties of magnetism, singles out spintronics as an important and very active research field. The success of the field depends in large on the design of materials and materials combinations that meet specific properties and functionalities, e.g. a maximum tunnel magnetoresistance ratio (TMR) at room temperature of a metal-oxide tunneljunction. It turns out that the computational materials design based on materials specific quantum mechanics, which means density functional theory (DFT) for most of the cases, combined with the computation of thermodynamical properties provides a successful path in stimulating experiments and guiding design principles. In this talk (i) we discuss the electronic structure of typical half-metals, of half-metals at surfaces and interfaces as obtained from ab initio calculations [1], (ii) we emphasize that interface states at the interfaces of tunneljunctions can have severe implications on the TMR [2], (iii) we show that the large TMR ratio of Fe/MgO/Fe is a result of a Bloch state filtering, which is sensitive to modifications of one atomic layer at the interface or small changes of the c/a ratio, (iv) we discuss the temperature dependence of the magnetic properties [3] by relating ab initio results to the classical Heisenberg model and thus investigating the Curie temperature of $MgO_{1-x}N_x$ in the context of a potential spin-filter material [4]. (v) We also propose a magnetic lead for metal/MgO/metal with a maximal Curie temperature above 1200 K [5] and an antiferromagnetically coupled TMR element made of half-metals without interface states based on an a priori understanding of the exchange interactions in such systems, as paradigms of materials design from first principles. The thermal properties of the Heisenberg model are determined by Monte Carlo calculations, while the long range Heisenberg exchange parameters are calculated on the basis of the density functional theory for example by performing spin-spiral calculations with finite cone-angles within the FLAPW method [6] or exploiting the Green function in the Korringa-Kohn-Rostoker Green function method [7]. Finally, we discuss results of the recently completed SPEX program [8], an efficient implementation of the GW approximation for the electronic self-energy within the all-electron FLAPW method as addition to our FLEUR project [9], which allows us to calculate the band gaps of oxides such as $MgO_{1-x}N_x$ beyond the local density approximation.

[1] M. Lezaic, Ph. Mavropoulos, G. Bihlmayer and S. Blügel, J. Phys. D: Appl. Phys. **39**, 797 (2006)
[2] M. Lezaic, Ph. Mavropoulos, J. Enkovaara, G. Bihlmayer and S. Blügel, Phys. Rev. Lett. **97**, 026404 (2006)
[3] Ph. Mavropoulos, M. Lezaic, and S. Blügel, Phys. Rev. B. **72**, 174428 (2005)
[4] Ph. Mavropoulos, M. Lezaic, and S. Blügel, submitted to Phys. Rev. B (arXiv:0908.0934v1)
[5] M. Lezaic, Ph. Mavropoulos and S. Blügel, Appl. Phys. Lett. **90**, 082504 (2007)
[6] Ph. Kurz, F. Forster, L. Nordstrom, G. Bihlmayer, and S. Blügel, Phys. Rev. B **69**, 024415 (2004)
[7] N. Papanikolaou, R. Zeller, P. H. Dederichs J. Phys.: Condens. Matter **14**, 2799 (2002); H. Ebert and R. Zeller, The SPR-TB-KKR package
http://olymp.cup.uni-muenchen.de/ak/ebert/SPR-TB-KKR
[8] C. Friedrich, S. Blügel, A. Schindlmayr, to be published
[9] http://www.flapw.de

Density functional theory and beyond with FHI-aims: Concepts and applications

Volker Blum
Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin

We describe the Fritz Haber Institute ab initio molecular simulations (FHI-aims) computer program package, an efficient computer code for all-electron quantum-mechanical simulations of molecules and solids, based on compact, accurate numeric atom-centered quantum-mechanical basis sets. The primary production method for total energies and gradients is density functional theory (LDA and GGA) for molecules and periodic systems. Additional functionality includes quantum-chemical approaches (Hartree-Fock, hybrid functionals, MP2) and self-energy-based methods (e.g., GW) for electronic spectroscopic properties. FHI-aims allows fully quantum-mechanical simulations for systems up to thousands of atoms, and performs efficiently also on massively parallel platforms (BlueGene) with possibly thousands of CPUs. We here demonstrate the approach (i) by tackling the formation of secondary structure in benchmark polyalanine peptides from first principles, and (ii) by a direct density functional theory assessment of the large-scale "hex" surface reconstructions of the Au(100) and Pt(100) surfaces, regarding structure, energetics, and a possible underlying electronic driving mechanism.

First-principles study of the multiternary semiconductors and their alloys

Shiyou Chen and Xin Gao Gong
Physics Department, Fudan University, Shanghai
Aron Walsh and Su-Huai Wei
National Renewable Energy Laboratory, Golden, CO, USA

First-principles calculations play an important role in the study of semiconductor alloys during the past 30 years, however, when the number of composition elements increases in multiternary semiconductors, it’s still a challenge to search for the most stable crystal structure among the large amount of possible configurations. Especially for the multiternary compounds containing neighboring elements in the periodic table, the experimental X-ray diffraction fails to determine their exact atomic configuration since the form factor of these atoms are similar, and hence we need to do theoretical search to make it clear. The octet rule, which states that atoms tend to combine in such a way that they lose or share electrons to have eight electrons in the valence shells, was usually taken as a guideline in the search for the low-energy III-V and II-VI semiconductor alloys. Here we show two example systems in which the octet rule CAN and CAN NOT be used as the guideline.

The first system is the zinc-blende chalcogenide semiconductors. We studied the sequential cation mutation in binary II-VI, ternary I-III-VI2 and quaternary I2-II-IV-VI4 compounds, and found that their crystal structures obey the octet rule and show an inheritance relation, i.e., the crystal structure of quaternary compounds can be derived from their ternary corresponding, and their structural parameters and band structure characters are also similar as those of the ternary compounds. Comparing with the experiments, we propose that: (i) most I2-II-IV-VI4 compounds are more stable in the kesterite structure, rather than the widely-recognized stannite structure; (ii) Cu and Zn layers are easy to be randomized in kesterite Cu_2ZnSnS_4 and $Cu_2ZnSnSe_4$, which makes the kesterite structure has the same symmetry as stannite. The close atomic number of Cu and Zn and their partial randomization are the main reasons for the previous structure confusion.

The second system is the superhard BC_2N and BC_4N, alloys of diamond and c-BN. Previous calculations show that structures obeying the octet rule even have higher energy than some other structures. In this system we proposed quantitatively a bond counting rule stating that structures with more C-C and B-N bond have lower energy, and based on this rule we performed an unconstrained structure search and identified a series of BN/C_2 (111) superlattices as the lowest-energy and most-compact structures. The calculated mechanical properties including bulk and shear modulus and ideal strengths also show that these (111) superlattices may have higher hardness than c-BN, second only to diamond. Due to the violation of the octet rule and the existence of polar interfaces, a strong internal electric field is observed in the long period BC_2Nn×n (111) superlattices, which explains the significant band gap decrease as the period n increases.

S. Chen, X.G. Gong, A. Walsh, and S.H. Wei, Appl. Phys. Lett. **94**, 041903 (2009)
S. Chen, X.G. Gong, A. Walsh, and S.H. Wei, Phys. Rev. B **79**, 165211 (2009)
S. Chen, X.G. Gong, and S.H. Wei, Phys. Rev. Lett. **98**, 015502 (2007)

Realistic description of the strongly correlated organics

Andreas Dolfen
German Research School for Simulation Sciences, Jülich

The physics of strongly correlated materials is one of the grand-challenges in condensed-matter sciences. Density functional theory fails due to the importance of the Coulomb repulsion between localized electrons. Instead we use non-perturbative many-body techniques which are, however, computationally demanding and only feasible for quite small model systems. We replace the full Hamiltonian of a real material with a model Hamiltonian comprising only the most important electronic degrees of freedom. The effect of all other electrons can merely be included in an average way in form of parameters. In the talk we will cover how to obtain these effective parameters using the one-dimensional organic metal TTF-TCNQ as an example. Moreover we show how we solve the resulting Hamiltonian with a parallel Lanczos method making efficient use of the memory and computational power of the massively parallel IBM BlueGene/P system for such calculations. To gain high-resolution angular-resolved spectral functions we employ cluster perturbation theory (CPT) which helps identifying signatures of spin-charge separation also found experimentally in TTF-TCNQ.

L. Cano-Cortès, A. Dolfen, J. Merino, J. Behler, B. Delley, K. Reuter, and E. Koch,
Eur. Phys. J. B **56**, 173 (2007)

Spin-orbit coupling in carbon-based nanostructured materials

Jian Zhou[1,3], Qifeng Liang[3], and **Jinming Dong**[2,3]
[1] Department of Material Science and Engineering, Nanjing University
[2] Department of Physics, Nanjing University
[3] National Laboratory of Solid State Microstructures, Nanjing University

The spin-orbit coupling (SOC) of carbon-based nanostructured materials has been studied using both the first-principles and tight-binding (TB) methods. We have found that: 1) The curvature-induced σ-π coupling in the single-walled carbon nanotube (SWNT) always causes a stronger SOC in their π-bonding states than their π^*-antibonding ones. And a microscopic mechanism has been proposed to satisfactorily explain the experimental observation that the SOC-induced band splitting in the SWNTs is different for electrons and holes [Nature 452, 448 (2008)], which can not be accounted for by the present theories. 2) The SOC-induced band splittings near the Fermi energies of the hydrogenated and fluorinated graphenes can be significantly enhanced to 10^{-2} eV from the original about 10^{-6} eV of the pure graphene, which is comparable to those found in the diamond and archetypal semiconductors. This huge SOC enhancement is found to be come not only from the sp^3 hybridization of carbon atoms, but also from the larger intrinsic SOC of the fluorine atom than the carbon one. We hope a lot of interesting phenomena caused by the SOCs (e.g. the spin Hall effect) can be experimentally observed in these systems.

Recent progress on linear scaling methods for electronic structure analysis

Weinan E
Princeton University and Peking University

The speaker will review some of the recent progress both on developing and on understanding linear scaling methods for electronic structure analysis. In the first half of the talk, the focus will be on subspace iteration algorithms for insulators. In the second half of the talk, the focus will be on metallic systems.

LDA+Gutzwiller Method for Correlated Electron Systems

Zhong Fang
Institute of Physic, Chinese Academy of Science, Beijing

We will introduce our newly developed ab initio LDA+Gutzwiller method, in which the Gutzwiller variational approach is naturally incorporated with the density functional theory (DFT) through the 'Gutzwiller density functional theory (GDFT)' (which is a generalization of original Kohn-Sham formalism). This method can be used for ground state determination of electron systems ranging from weakly correlated metal to strongly correlated insulators with long-range ordering. We will show that its quality for ground state is as high as that by dynamic mean field theory (DMFT), and yet it is computationally much cheaper. In addition, the method is fully variational, the charge-density self-consistency can be naturally achieved, and the quantities, such as total energy, linear response, can be accurately obtained similar to LDA-type calculations. Applications on several typical systems will be presented, and the characteristic aspects of this new method will be demonstrated. Finally, by applying this method to FeAs-based compounds, we will show that some of the difficult issues, such as the Fe-As bond length and the magnetic moment of the SDW state, can be solved.

[1] X. Y. Deng, X. Dai, Z. Fang, Europhys. Lett. **83**, 37008 (2008)
[2] G.T. Wang, X. Dai, Z. Fang, Phys. Rev. Lett. **101**, 066403 (2008)
[3] X.Y. Deng, L. Wang, X. Dai, Z. Fang, Phys. Rev. B **79**, 075114 (2009)

Predictive Modeling of Novel Structures and Structural Phase Transitions in Zero and One Dimensions

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Fudan University, Shanghai

Nanostructured materials usually have different physical properties from their bulk phases and show strong dependence on size with well-defined scaling behavior. However, when the sizes become too small, deviations from such scaling properties may emerge, as illustrated in the present talk using a few intriguing examples of metal nanoclusters and nanowires. We show, based on first-principles calculations, that strong relativistic effects can make gold clusters to be cage-like, which is non-compact and similar to C_{60}. Strong magnetic interaction can enhance the stability of layer-like structure of small Co clusters, similar to the fragment of bulk phase. For nanowires, we show how the size effect will lead to a peculiar cross-section that is in contrast with the prediction of the classic Wulff plot. I will also present examples illustrating how hydrostatic pressure induces structural transitions in nanoclusters and carbon nanotubes.

Continuous-time quantum Monte Carlo solvers for DMFT

Evgeny Gorelov
Institut für Festkörperforschung, Forschungszentrum Jülich

We discuss the numerically exact continuous-time quantum Monte Carlo (ct-QMC) algorithm in the weak coupling scheme for electronic structure calculations of strongly correlated multi-band models in the framework of dynamical mean-field theory (DMFT). As a example, we discuss the end members of the family of layered rithenates $Ca_{2-x}Sr_xRuO_4$. In particular, we analyze the nature of metal-insulator transition in Ca_2RuO_4. As a second application of the ct-QMC solver, we study single magnetic impurities in a non-magnetic environment (Kondo systems), particularly a Co impurity in/on Cu crystals.

Electron-phonon coupling in strongly correlated systems

Olle Gunnarsson
Max-Planck-Institut für Festkörperforschung, Stuttgart

The electron-phonon interaction (EPI) is discussed for strongly correlated systems. For non-interacting electrons, effects of the EPI can often be characterized by the coupling strength and the phonon frequency. For strongly correlated systems this information is not sufficient. It is argued that effects of the EPI depends crucially on which phonons are considered and which property is studied. These issues are discussed, using examples from alkali-doped fullerides and high-Tc cuprates.

Method to construct transferable minimal basis sets for ab initio calculations

Lixin He
Key Laboratory of Quantum Information, University of Science and Technology of China, Hefei

We propose a new scheme to construct transferable minimal basis of localized orbitals for ab initio calculations. We first extract a set of highly localized Wannier-like orbitals from the reference systems. For each orbital, we decompose it to a pseudo-atomic orbital, augmented by small local functions centered at its nearest neighbor atoms. When applied for a real system, the center of each local function moves with its associated atoms, without changing its shape and amplitude. We have done intensive tests of this scheme for III-V and group IV semiconductors and find it has very good transferability, way better than the original Wannier-like orbitals, while still keep the basis size minimal. This work sheds new lights on why Wannier and Wannier-like functions are not transferable as one may expect.

Understanding spin-sensitive scanning probe microscopy based on electronic structure theory

Stefan Heinze
Institut für Theoretische Physik und Astrophysik, Universität Kiel

In recent years, spin-polarized scanning tunneling microscopy (SP-STM) has been established as a technique to resolve magnetic nanostructures down to the atomic scale [1-4]. Very recently, it has even become possible to measure the exchange interactions between tip and sample by magnetic exchange force microscopy (MExFM) [5,6]. However, the interpretation of such measurements is non-trivial, especially on the atomic scale. For SP-STM there are various contributions to the tunneling current from structural, electronic, chemical, and magnetic sample properties. For MExFM there are many basic open questions concerning e.g. the influence of tip material, structure, and sample systems on the size, sign, and distance dependence of exchange forces. Successful interpretation approaches for both techniques rely on an accurate description of the electronic structure of tip and sample and their interaction making density functional theory (DFT) an indispensable tool.

Here, I will present the theory of SP-STM and explain how one can reveal collinear and non-collinear magnetic order on the atomic scale [1-4]. This allows us to verify theoretical predictions based on DFT such as two-dimensional antiferromagnetism [1] or spin-spiral structures driven by the Dzyaloshinskii-Moriya interaction [3,4]. Surprisingly, it is even possible to image non-collinear atomic-scale magnetic order using a non-magnetic tip as the electronic structure changes slightly from atom to atom due to spin-orbit coupling. Finally, I will show how the antiferromagnetic order of Fe on W(001) can be resolved by MExFM and discuss how the structure and chemical composition of the tip influence the measured forces [6].

[1] A. Kubetzka et al., Phys. Rev. Lett. **94**, 087204 (2005)
[2] K. von Bergmann et al., Phys. Rev. Lett. **96**, 167203 (2006)
[3] M. Bode et al., Nature **447**, 190 (2007)
[4] P. Ferriani et al., Phys. Rev. Lett. **101**, 027201 (2008)
[5] U. Kaiser et al., Nature **446**, 522 (2007)
[6] R. Schmidt et al., Nano Lett. **9**, 200 (2009)

Ab initio molecular dynamics for structural properties of layered Mott insulators and iron pnictides

Harald O. Jeschke
Institut für Theoretische Physik, Goethe-Universität Frankfurt am Main

We employ Car-Parrinello molecular dynamics for the investigation of the structural response of low dimensional materials under the influence of pressure or doping. For the multiorbital Mott insulator TiOCl, we study the pressure induced transition to a metallic phase. This phase is characterized by a strong Ti-Ti dimerization. At even higher pressures, we predict the dimerization to disappear in a second phase transition. In an investigation of the effect of sodium intercalation of the layered material TiOCl, we predict the positions of the sodium ions in superlattice structures and show that surprisingly Na fails to metallize the Mott insulator TiOCl for all studied concentrations. We propose a description in terms of a multiorbital ionic Hubbard model.

For the AFe_2As_2 (A=Ca, Sr, Ba) family of iron pnictide superconductors, we we investigate the electronic and lattice structure under pressure. We observe that the structural phase transition from orthorhombic to tetragonal symmetry is always accompanied by a magnetic phase transition in all the compounds, while the nature of the transitions is different for the three systems. Our calculations explain the origin of the existence of a collapsed tetragonal phase in $CaFe_2As_2$ and its absence in $BaFe_2As_2$. Our results show the large potential of unbiased structural relaxations using ab initio molecular dynamics for the study of pressure and doping induced effects.

Molecular dynamics/density functional (MD/DF) calculations for phase change memory materials : progress and promise

Robert O. Jones
Institut für Festkörperforschung, Forschungszentrum Jülich

The latest generation of supercomputers has changed dramatically our ability to perform calculations on materials, and we shall demonstrate this for phase change materials (PCM). These are the recording layers of familiar rewriteable optical storage devices such as CD, DVD, and Blu-ray Disc. Nanosize amorphous bits are "written" by a laser in a polycrystalline layer and "erased" by another laser pulse in a crystallization process at a temperature below the melting point. Understanding such processes is impossible without knowing the structures of the phases involved, and DF calculations are ideal for this purpose. We show that MD/DF calculations are possible for hundreds of atoms over a time scale approaching those relevant experimentally. Our examples include amorphous Ge/Sb/Te alloys, and we show that the results can be used in conjunction with experimental data to provide convincing structural details.

J. Akola, R. O. Jones et al., Phys. Rev. B **80**, 020201(R) (2009)

Quantum cluster extensions of dynamical mean-field theory

Erik Koch
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Dynamical mean-field theory (DMFT) describes a correlated solid as a single unit-cell embedded in a self-consistent bath. A natural generalization is to increase the number of lattice sites treated explicitly, i.e. considering clusters. The DMFT approximation to the self-energy is then less severe. Moreover on a cluster it is possible to study (short-range) correlations, which are not accessible in a single site approximation. Cluster extensions of DMFT are, however, not unique. We show that the two common approaches, cellular DMFT (CDMFT) and the dynamical cluster approximation (DCA), are merely two particular choices of a general gauge freedom, the phase on the cluster sites.
As a test case we consider the one-dimensional Hubbard chain, for which dynamical mean-field theory should work least well. We compare the results of DMFT calculations for increasing cluster size to the exact results and discuss the non-locality of the self-energy close to the Mott transition.

E. Koch, G. Sangiovanni, O. Gunnarsson, Phys. Rev. B **78** 115102 (2008)

Electrocatalysis: Going to atomic level

Zhi-Pan Liu
Laboratory for the Computational Physical Sciences, Ministry of Education, and
Department of Chemistry, Fudan University

In this talk, I will introduce some general background of electrocatalysis and the current difficulty in theoretical simulation. Some of the recent theoretical development in the world and our group will be presented. Based on our recent studies on fuel cell applications, the factors affecting the electrocatalytic kinetics will be summarized. We suggest that the electrical potential itself is not the key factor that directly affects the reaction barrier of surface reactions. Instead, the surface coverage and the morphology can be much more important.

H.F. Wang and Z.P. Liu, J. Am. Chem. Soc. **130**, 10996 (2008)
Y.H. Fang; Z.P. Liu, J. Phys. Chem. C **113**, 9765 (2009)

Boron and boron-carbon nanostructures

Jun Ni
Department of Physics, Tsinghua University, Beijing

Low dimensional structures, such as clusters, planar sheet and nanotubes, often show striking properties with various applications in electronic and magnetic nano-devices. For carbon nanostructures, there are C_{60} fullerene cage, graphene and carbon nanotubes, which have the similar constructed units due to the sp^2 hybridization. As the nearest-row neighbor of carbon, boron is fascinated for the multicentre bonds in the crystal structures. In this talk, I will report our recent theoretical results of boron and boron-carbon nanostructures. The boron sheet is metal. Rolled from the sheet, the nanotubes can be either metals or semiconductors dependent of diameter and chirality, which may exhibit potential applications similar to carbon nanotubes. The boron nanoribbons constructed from the stable boron sheet also shows rich electronic properties. The bare boron nanoribbons with different edges are metals, while the boron nanoribbons with hydrogen passivated on the zigzag edges become semiconductors. Boron and carbon can form binary ordered nanostructures. The BC_3 nanoribbons with the armchair shaped edges are all semiconductors, while BC_3 nanoribbons with the zigzag shaped edges are semiconductors or metals dependent on the edge atoms. I will also discuss the hydrogenation of boron-carbon ordering sheets. We find that hydro-boron-carbon have both stable semiconducting and metallic conformations. Because of the rich electronic properties, the boron-carbon nanostructures are the potential low-dimensional materials for nano-electronic and device applications.

Origin of orbital-order in $KCuF_3$ and $LaMnO_3$

Eva Pavarini
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Orbital-ordering phenomena play a fundamental role in determining the electronic and magnetic properties of many strongly correlated oxides. The origin of orbital-order in real materials is, however, still controversial. The classical examples of orbitally-ordered systems are $KCuF_3$ and $LaMnO_3$, two materials with very different physics. Using the LDA+DMFT approach we disentangle the effects of the different lattice distortions, electron localization, and many-body superexchange and clarify the mechanism of orbital-order in these two paradigmatic compounds.

E. Pavarini, E. Koch, and A.I. Lichtenstein, Phys. Rev. Lett. **101**, 266405 (2008)
E. Pavarini and E. Koch, arXiv:0904.403

First principle calculation on the vacancy formation on TiO_2 surface

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The most common defects on surfaces of transition metal oxides are oxygen vacancies, which play critical roles in applications such as heterogeneous catalysis, photoelectrolysis, biocompatibility, etc.. If the nature and distribution of the oxygen vacancies can be controlled, the surface properties will then be modified for different applications. For this purpose, one needs to understand both the influence of oxygen vacancies on the surface properties and the responses of oxygen vacancies to different external fields. We have conducted comprehensive first principles calculations on the surface energy of strained rutile TiO_2(110) with oxygen vacancies. The formation energy of each type of oxygen vacancy is calculated as a function of external strain. We find that the type of the most easily formed oxygen vacancy can be tuned by the strain and therefore suggest that the distribution of oxygen vacancies can be engineered by external strain, which helps to improve the applications of TiO_2 surface where oxygen vacancies play important roles. The dependence of surface elastic properties on the type of oxygen vacancy is found to be responsible for the interplay between external strain and oxygen vacancies.

D.J. Shu, S.T. Ge, M. Wang and N.B. Ming, Phys. Rev. Lett. **101**, 116102 (2008)

Capillary force-induced structural instability in liquid encapsulated elastic circular tubes

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The capillary-induced structural instability of an elastic circular tube partially filled by a liquid is studied by combining theoretical analysis and molecular dynamics simulations. The analysis shows that, associated with the instability, is a well-defined length scale (elasto-capillary length), which exhibits a generic scaling relationship with the characteristic length of the tube, regardless of the interaction details. We validate this scaling relationship for a carbon nanotube partially filled by liquid iron. The scaling relation and instability found in this study have potential applications for nano-devices.

Computational design of materials for hydrogen storage in quasi-molecular form

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Hydrogen is the least complex and most abundant element in the universe. As an energy carrier, hydrogen is non-toxic, renewable, clean to use, and packs much more energy per pound. Therefore hydrogen will play a critical role in a new, decentralized energy infrastructure that can provide power to vehicles, homes, and industries. However, one of the biggest challenges in a new hydrogen economy is storage, since the storage medium must meet the requirements of high gravimetric and volumetric density, fast kinetics and favorable thermodynamics. The current methods of storing hydrogen as compressed gas or in the liquid form does not meet the industry requirements since the energy densities are much lower than that in gasoline. Moreover, there are issues of safety and cost involved in compressing hydrogen under high pressure or liquefying it at cryogenic temperatures. Although storage of hydrogen in solid state materials offers an alternative, there are no current solid state storage materials that meet the industry requirement. By using state-of-art simulation techniques, we have systematically studied the interactions of hydrogen molecules with diverse nanostructures in order to gain insight into the design of new materials for hydrogen storage. In this talk I will review our research progress in computational design of nanomaterials for hydrogen storage, focusing on (1) Functionalized fullerenes and heterofullerenes for hydrogen storage; (2) Coordinately unsaturated metal sites in matrix for hydrogen storage; (3) hybrid porous materials for hydrogen storage. These subjects stand for the current new directions in searching for new materials for hydrogen storage.

Calculated magnetic exchange interactions in high-temperature superconductors

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Using a first-principles linear-response approach, we study the magnetic exchange interactions *J* for a series of superconducting cuprates. We reproduce the observed spin-wave dispersions together with other experimental trends, and show that different cuprates have similar *J*'s regardless of their T_c. The nearest-neighbor *J* is not sensitive to the hole doping, which agrees with recent experiments. For the undoped cuprates, the second nearest-neighbor *J* is ferromagnetic, but changes its sign with hole doping. We also find that, in contrast to the hopping integral, the exchange interaction is not sensitive to the position of apical oxygen. To see the effect of the long-range nature of the exchange on the superconducting T_c, we study the dynamical spin susceptibility $\chi(q,\omega)$ within the *t*-*J* model using a dynamical cluster approximation.

Computational Study of Unusual Material Properties of Oxides

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Post-transition metal oxides (ZnO, In_2O_3, SnO_2, etc.) and their alloys comprise a group of materials that have many unique physical properties such as simultaneously high electron conductivity and optical transmission, high catalytic efficiency, and chemical stability. Therefore, they are widely used in optoelectronic applications such as photovoltaic and photoelectrochemical devices, LEDs, display panels, and chemical sensors. They have also been proposed as ideal hosts for high TC spintronic materials.

Using first-principles method, we have calculated material properties of the metal oxides, demonstrating that the properties of the binary and multinary oxides are more complex than previously thought. We find that (i) many oxides (In_2O_3, Cd_2SnO_4, etc.) have non-equivalent fundamental and optical band gaps, arising from parity forbidden band edge transitions; a good transparent conducting oxide should have small fundamental band gap but a large optical band gap [1]. (ii) The band gap renormalization in degenerate n-type doped oxides arises from the nonparabolic nature of the conduction band (i.e., not a rigid shift as previous thought), and is highly sensitive to the electronic states of the dopants [2]. (iii) The ground state crystal structure of the non-isostructural $(InMO_3)_m(ZnO)_n$ (M=In, Ga, Al) alloy should satisfy several fundamental rules such as the octahedron rule and the octet rule [3]. The formation of the layer quaternary structure or amorphous structure also results in some interesting changes in the electronic and optical properties of the alloy [4]. (iv) The origin of the electron and hole induced ferromagnetism in oxides are investigated [5, 6]. For example, we show that the spontaneous magnetization in d^0 oxides with sufficient holes is intrinsic properties of the oxides and can be enhanced by anion site doping or through quantum confinement [6]. (v) We have proposed a designing principle for the band edge modification of the oxide materials. For example, we demonstrate that TiO_2 co-doped with Mo and C is suitable for photoelectrochemical water splitting applications [7]. (vi) Finally, we have analyzed the doping asymmetry problem in the wide-gap oxides and proposed several approaches to overcome the p-type and bipolar doping bottlenecks in these materials [8].

[1] A. Walsh, J. L. F. Da Silva, S.-H. Wei et al., Phys. Rev. Lett. **100**, 167402 (2008)
[2] A. Walsh, J. L. F. Da Silva, S.-H. Wei, Phys. Rev. B **78**, 075211 (2008)
[3] J. L. F. Da Silva, Y. Yan and S.-H. Wei, Phys. Rev. Lett., **100**, 255501 (2008)
[4] A. Walsh, J. L. F. Da Silva, Y. Yan, M.M. Al-Jassim and S.-H. Wei, Phys. Rev. B **79**, 073105 (2009)
[5] A. Walsh, J. L. F. Da Silva, S.-H. Wei, Phys. Rev. Lett. **100**, 256401 (2008)
[6] H. Peng, H. J. Xiang, S.-H. Wei et al., Phys. Rev. Lett. **102**, 017201 (2009)
[7] Y. Gai, J. Li, S.-H. Wei et al., Phys. Rev. Lett. **102**, 036402 (2009)
[8] Y. Yan, J. Li, S.-H. Wei et al., Phys. Rev. Lett. **98**, 135506 (2007)

Determination of structural and magnetic properties of dilute magnetic semiconductors with x-ray magnetic circular dichroism

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We will report our recent findings that x-ray magnetic circular dichroism (XMCD) spectra of 'nonmagnetic' atoms or the k-edge XMCD spectra of magnetic dopants strongly depend on the local atomic arrangement and can hence be used for the structural characterization for dilute magnetic semiconductors and their interfaces. As an example, we found that besides single Co ions on substitutional sites, there are also a large amount of interstitial Co atoms and pairs of Co-oxygen vacancy in the as-grown Co-doped ZnO sample. Comparisons between density functional results and experimental data for Mn-doped GaAs and Fe/GaAs(100) interfaces will also be discussed.

Nearly Free Electron States in Gated Graphene Nanoribbon Superlattices

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We propose a facile way to obtain one-dimensional electron gas (1DEG) in vacuum by occupying nearly free electron (NFE) states in graphene nanoribbon (GNR) superlattice, which can be used to construct graphene based field effect transistor. Our first principles calculations suggest that there are two types of NFE states in GNR superlattices. One type locates close to ribbon surfaces while the other type locates in the vacuum between two ribbons. Upon electron doping, the latter is easy to be lowered in energy and even be occupied, forming a clean 1DEG system. These two types of NFE states can be simply interpreted using a 1D Kronig-Penney potential model.

First principles investigation on quantum transport of atomic and molecular wires

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We studied quantum transport of carbon atomic wires and alkane molecules sandwiched between gold electrodes using a first-principles method based on density functional theory and nonequilibrium Green's function formalism. We found finite sized atomic wires with even number of carbon atoms have magnetism, while wires with odd number atoms have no net magnetic moment. Perfect spin filtering effect can be obtained by slightly straining the wire after it was connected to electrodes. We also explored the effects of binding modes and anchoring groups on electronic transport properties of alkane chains. When the alkanes anchored with SH, NH_2, or COOH groups, all the conductance values related to the four binding modes considered are found to divide into two sets: high and low conductance, explaining well the observations in recent experiments. When the bias increases to non-linear *I-V* region, the characteristic of two sets of the conductance is found to disappear. The results may be useful for the application of atomic and molecular wires in molecular electronics.

Characterizing graphene by doping

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Electronic structure of zigzag-edged graphene nanoribbons (ZGNRs) doped with boron(B) or nitrogen(N) atoms are investigated by spin polarized first-principles calculations. Interesting phenomena have been inspected in the doping systems, such as the alternation between semiconducting, half-metallic or metallic by controlling the distance of the impurity atoms to the edges, spin filtering effect in doped graphene device, and the enhancement of lithium storage capacity in boron doped graphene; etc. The mechanisms of these effects are discussed and our results have provided useful approaches to manipulate the graphene.

X.H. Zheng, G.R. Zhang, Z. Zeng, V.M. Garcia-Suarez, and C.J. Lambert, Phys. Rev. B **80**, 075413 (2009)
X.H. Zheng, R. Wang , L. Song , Z.X. Dai , X.L. Wang , Z. Zeng, APL, accpepted
X.H. Zheng, X.L. Wang, T.A. Abtew and Z. Zeng, PRB, submitted
X.L. Wang, Z. Zeng, and G.X. Wang, APL, submitted

Finite Element Method for Solving Kohn-Sham Equations Based on Self-adaptive Tetrahedral Mesh

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We have developed a parallel FE solver for the Kohn-Sham equation based on the combination of density functional theory and pseudopotential. FE method is a typical real-space method with strictly local basis function, it produces the sparsely structured Hamiltonian matrices, does not require Fourier transforms and well suits for parallel implementation. In addition, it is relatively straightforward for implementing adaptive refinement techniques to pay more attention to the regions where the computed functions vary rapidly, such as regions around atomic nuclei or chemical bonds. Therefore, the accuracy of the calculation can easily be improved by refining the mesh. We will show how the FE nodes can be distributed by fitting the error of the wave-functions, so-called self-adaptive refinement. The efficient eigenvalue solver for the large-scale sparse matrix in parallel calculation is another challenge. Conjugate gradient (CG) is a powerful method in electronic-structure calculation what benefits from the initial-vectors in SCF iteration. We found an effective preconditioner for the CG method especially in the un-regular FE mesh.

Methodology and capabilities of the KKR Green function method

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An introduction is given to the full-potential Korringa-Kohn-Rostoker Green function method, in particular to the version which has been developed in Jülich for density functional calculations of bulk systems, surfaces and impurity atoms in bulk systems and at surfaces. The conceptual difference of the KKR Green function method compared to the standard basis set approach is discussed and it is shown that lattice constants and bulk moduli for metals and semiconductors and forces and displacements in crystals perturbed by impurities can be determined in very good agreement with full-potential linear augmented plane-wave (FLAPW) and plane-wave pseudo-potential calculations. A transformation into a tight-binding (TB) form (essentially Andersen's screened KKR method) is explained which leads to linear-scaling computational effort for layered systems like slabs, surfaces, interfaces and semi-infinite crystals and which can be used for bulk systems with thousands of atoms if the KKR matrix equations are solved by iteration.

Participants

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