Ψ_k Newsletter

AB INITIO (FROM ELECTRONIC STRUCTURE) CALCULATION OF COMPLEX PROCESSES IN MATERIALS

Number 84 December 2007

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Contents

1 Editorial

In this last newsletter of this calendar year, we would like to turn readers attention to four items. First, we would like to recommend two reports, one on the ESF Workshop Symposium for Theoretical Chemistry - Large, Non-biological Systems, and the second a UK CCP9 Workshop on ”Solving Bogoliubov-de Gennes and Gross-Pitaevskii Equations for Superconductors, Superfluids and BEC”. Both reports contain abstracts of presented papers. The next interesting item is the announcement of the ”SaX” code, introduced briefly by its developers in the section ’Presenting Other Initiatives’. The last item which we would like to recommend is the scientific highlight of the month by Artem R. Oganov (Zurich and Moscow), Yanming Ma (Zurich and Jilin), Colin W. Glass (Zurich), and Mario Valle (CSCS Switzerland) on ”Evolutionary crystal structure prediction: overview of the USPEX method and some of its applications”. Further details on the newsletter can be found by inspecting the table of contents.

Regarding the ’Psi-k Portal’, it is still work in progress, and no e-mails are being distributed through the portal as yet. So, in order to receive all the Psi-k annoncements one still needs to join also the usual Psi-k mailing list. As soon as we switch fully to the Psi-k Portal, a message will be sent to the Psi-k mailing list with the information to this effect.

Finally, as it is the last newsletter of the year, we would like to take the opportunity and thank all the contributors for their submissions, and the readers for their interest. May the next year be succesful for everybody!

The *Psi-k Network* has a home page

http://www.psi-k.org

The above contains information on the present and future Psi-k workshops and hands-on courses (subject to funding).

Please submit all material for the next newsletters to the email address below.

The following email addresses, which remain in operation, are repeated for your convenience, and are the easiest way to contact us.

	function
psik-coord@dl.ac.uk	**messages to the coordinators, editor & newsletter**
psik-network@dl.ac.uk	**messages to the whole Ψ_k community**

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2 News from the ESF Programme

”Towards Atomistic Materials Design”

2.1 Reports on ESF Workshops

2.1.1 Report on Workshop Symposium for Theoretical Chemistry – Large, Non-biological Systems

University of Saarland, Saarbrücken, Germany

September 16th – 20th 2007

Sponsored by European Science Foundation, German Research Council, Fonds der Chemischen Industrie, Kontaktstelle für Wissens- und Technologietransfer der Universität des Saarlandes, Landeshauptstadt Saarbrücken, Saartoto, Ministerium für Bildung, Kultur und Wissenschaft (Saarland), Siemens, Sun Microsystems, Teubner Verlag, Vieweg Verlag, Universität des Saarlandes, Wiley-VCH

Organized by Michael Springborg (University of Saarland)

Web page: http://www.stc2007.uni-saarland.de

Symposium for Theoretical Chemistry – Large, Non-biological Systems

Report

The symposium for theoretical chemistry took place in Saarbrücken September 16th – 20th 2007 on the campus of the University of Saarland, Germany. It was organized by Michael Springborg (Saarbrücken) and his team (Michael Bauer, Anton Bayer, Yi Dong, Valeri Grigoryan, Hanno Kamp, Elisaveta Kasabova, Marco Schüle, Violina Tevekeliyska, Habib Ur Rehman, and Ingelore Weidenfeld). Enormous support with the organization was provided by Uta Merkle and her team at the *Kontaktstelle für Wissens- und Technologietransfer* (KWT) at the University of Saarland.

The informal opening of the conference took place at a reception Sunday evening in the townhall of the town of Saarbrücken. In the official opening, Monday September 17th, it was emphasized that the symposium had become internationally recognized, whereas the first such symposia were essentially limited to the German-speaking countries. With the main topic of the symposium, large, non-biological systems, it was possible to bridge a gap between physics and chemistry, although also contributions to smaller and/or biological systems were presented. During the following 3 1/2 days 12 invited and 24 contributed talks as well as almost 170 posters were presented. The conference was organized as a low-cost conference and it was encouraging to see that many young scientists considered that a possibility to attend and also present their work.

The first session was devoted to molecular-dynamics simulations. Dominik Marx (Bochum, Germany) reported about the latest developments in ab initio, molecular dynamics simulations, including presenting an approach for studying antiferromagnetic coupled spin systems. In the next two talks, Daniel Sebastiani (Mainz, Germany) reported about the calculation of momentum densities of protons and Petr Slavíček (Prague, Czech Republic) told about his studies on photochemical reactions of molecules and clusters.

The second session was opened by a talk by Richard Martin (Los Alamos, USA) who told about oxides of f electron elements. Theoretical studies of those systems are non-trivial due to strong correlation effects combined with relativistic effects. Also the presentation of Hélène Bolvin (Strasbourg, France) was devoted to heavy elements. She described an approach for calculating g matrix elements and applied it on various actinide compounds. In his talk, Anatoly Titov (St. Petersburg, Russia) discussed relations between the properties of heavy-atom molecules as calculated with 'normal' electronic-structure methods and those predicted by more fundamental approaches based on the standard model.

In his talk, John Perdew (New Orleans, USA) gave a clear overview of the 'Jacob's ladder' of density functionals, with increasingly advanced functionals on the higher rungs. LDA and GGA functionals form the first two rungs, and together with the meta-GGA functionals of the third rung they are all completely non-empirical constructions. The latest developments, the hyper-GGA functionals on the fourth rung, on the other hand, are based on an orbital-formulation of exchange energy, whereby a cancellation of errors between exchange and correlation functionals is lost and, simultaneously, one has to introduce empirical parameters. That he used 'old-fashioned' viewgraphs that even were modified 'on the fly' added to the interest that his talk arouse. In two further talks, by Kartick Gupta (University of Burdwan, India) and Laszlo von Szentpaly (Stuttgart, Germany), methods for using descriptors to study chemical acitivities of individual molecules were discussed.

Poul Jørgensen (Århus, Denmark) and Christian Ochsenfeld (Tübingen, Germany) presented methods that allowed for efficient theoretical treatments of very large systems. With these methods also larger fragments of a DNA molecule could be treated (with more than 1000 atoms and 10000 basis functions). Subsequently, Andreas Köhn (Mainz, Germany) discussed the transfer of excitation energies between donor and acceptor molecules. Andreas Köhn presented an accurate approach for the theoretical study of these processes.

In the last talk on the first day of the symposium, Benoît Champagne (Namur, Belgium) presented methods and results for the calculation of structural, mechanical, electronic, and optical properties of regular polymers. He showed that an approach based on periodicity is much more efficient than one based on finite systems of increasing size.

Monday evening was devoted to the first poster session. During some three hours halfpart of the posters was presented and intensively discussed. To the pleasure of the organizers, the larger part of the attendees stayed at the symposium also during the evening.

Tuesday morning was reserved for the presentation of the Hellmann award winner, Andreas Dreuw (Frankfurt/Main, Germany). After having received his award, he presented his work on photoinduced electron transfer in biochemical systems. Subsequently, Serguei Fomine (Mexico City, Mexico) presented theoretical predictions for the properties of tubular systems of oligothiophenes. In her talk, Alisa Krishtal (Antwerp, Belgium) analysed in detail methanol clusters with the ultimate purpose of understanding how the macroscopic properties of a solvent (i.e., methanol) depend on the microscopic properties of the solvent molecules.

Benedetta Mennucci (Pisa, Italy) discussed the most recent extensions of the polarizable continuum method for the study of molecules in solution. Within one extension, also excited states can be treated. She pointed out the srengths and limitations of the different methods for treating solvated molecules. Jan-Ole Joswig (Dresden, Germany) used a parameterized density-functional method in studying proton transport in polyphosphonic acids that are considered interesting materials for fuel cells. Dipole switching processes in large molecules was the topic of the presentation of Tillmann Klamroth (Potsdam, Germany).

Catalytic properties of surfaces of oxides were discussed by Joachim Sauer (Berlin, Germany). He demonstrated how a close collaboration with experimentalists can be very useful in obtaining a detailed understanding of the materials properties. Dynamical properties were studied by Judith Voll (Munich, Germany), who considered laser induced dynamics of β-carotene using a simplified model. Ozlem Ozcan (Düsseldorf, Germany) presented his results of Monte Carlo – density-functional studies of the adsorption of organosilanes on a ZnO surface.

Arne Lüchow (Aachen, Germany) discussed advantages (accuracy) and disadvantages (limited size of the treatable systems) of quantum Monte Carlo simulations and gave also examples of his most recent applications of this approach. Chemical reactions was the topic of the presentation of Vincent Tognetti (Paris, France) who studied a catalytic process of ethylene dimerization. Philipp Scherer (Munich, Germany) exploited in detail a simple model system for the study of electron transfer processes in large biological systems.

Before the second poster session, Frank-Dieter Kuchta of the German Research Council (DFG) gave a very useful overview of the activities of DFG and of the different programs of this funding agency. After the dinner, the second halfpart of the posters was presented, and, as at the first poster session, this poster session was very well attended.

In the first talk on Wednesday morning, Karlheinz Schwarz (Vienna, Austria) presented the latest developments of the WIEN2k computer programs for the study of crystalline materials. This included also recent applications, where, e.g., the LDA+U approach was used to get a proper description of the materials. By combining information from more different types of experiment, the value of the parameter U could be settled quite accurately. In another application, he showed that also surfaces can be studied. This included an impressive example of a borazine nanomesh on the Rh(111) surface whose structure was accurately determined. A perfect agreement with experimental observables was obtained, although the interpretation differed. This system contained about 1100 atoms per unit cell.

David Benoit (Ulm, Germany) presented a new method for the calculation of vibrational properties of large systems. Since these properties are important for the characterization of the systems, such theoretical developments are very useful in providing links between theory and experiment. Liviu Hozoi gave results for the study of extended systems were correlation effects were added with the so called local methods to a Hartree-Fock based calculation.

The second morning session on Wednesday was devoted to larger, but finite systems. First, David Wales (Cambridge, UK) discussed ways of describing and determining the 'energy landscapes' of such systems, i.e., the total-energy hypersurfaces. The so called disconnectivity graphs provide a useful two-dimensional presentation of the hypersurfaces that also allow for analysing how easily a local or global total-energy minimum can be reached. Subsequently, Konstantin Neyman (Barcelona, Spain) and Christoph Scheurer (Munich, Germany) discussed the properties of different specific nanoparticles.

Wednesday afternoon was devoted to an excursion were a couple of interesting places in Saarland were visited. These were the Saar Loop where the river Saar makes a spectacular 180° turn as well as the iron work in Völkingen that is a Unesco World Cultural Heritage. The day closed with a conference dinner at the university.

The last day of the symposium, Thursday, started with a presentation by Martin Kaupp (Würzbung, Germany) who shared his time between two topics, the theoretical determination of NMR parameters and the development of new hybrid density functionals where the hybrid parameter was allowed to be position dependent. In his talk, Dominik Kröner (Potsdam, Germany) presented a theoretical study of the laser control of the switching of a chiral molecule. Due to the complexity of the problem at hand, the calculations were based on a simplified model. Subsequently, Wolfgang Eisfeld (Bielefeld, Germany) discussed the results of extensive calculations on a small molecule, NO_3, where also all vibrational degrees of freedom were included in the dynamical simulations.

The last session of the symposium was devoted to crystalline compounds. Hermann Stoll (Stuttgart, Germany) discussed his approach for splitting correlation into a long- and a short-ranged part that can be treated separately. The short-ranged parts are treated with density functional methods, and the long-ranged parts with wavefunction based methods. Ultimately, this leads to a highly accurate method. Beate Paulus (Dresden, Germany) applied the method of local increments for the inclusion of correlation effects to study the adsorption of CO on a ceria surface. Finally, Ulrich Wedig (Stuttgart, Germany) demonstrated that even some crystal structures of some elements (Zn and Cd) are not well understood.

In total the conference demonstrated that the field of computational materials science / chemistry / physics is very active and that methodological developments have made it possible to apply computational approaches to the study of systems and situations that accurately match those found in experiment, and not only idealized model system. Thereby, such calculations have become truely predictive. Although also the development in computer technology has been of paramount importance, the most significant improvements have been obtained through improved computational and theoretical methods.

The developments that have made it possible to use computational approaches predictively include the treatment of dynamical properties of larger systems, methods for studying systems in realistic atmospheres and solutions, the study of chemical reactions in gas phase and on catalytic surfaces, and the possibility of studying structural and electronic properties of large, but finite, nanosystems. But also the development of highly accurate density functionals in density-functional theory and of alternative methods for studying certain systems at a more accurate level remains being very important for assuring that results obtained with theoretical methods become increasingly accurate and reliable.

The systems that were presented during the symposium represented an impressive range from crystalline or biological systems with more than 1000 atoms being treated explicitly, to the study of dynamical properties in materials of relevance for the development of fuel cells. The calculation of all kinds of response functions provided a close link to experimental studies.

Finally, further information on the symposium, including pictures taking during the 3 1/2 days, can be found on the homepage of the symposium, http://www.stc2007.uni-saarland.de, at least until the end of the year 2007.

Saarbrücken, October 2007, Michael Springborg

Programme

Sunday, September 16th

Reception

17.00 – 20.00 Registration in the Town Hall of the Town of Saarbrücken

18.00 – 20.00 Reception in the Town Hall of the Town of Saarbrücken

Monday, September 17th

From 07.45 Registration in front of the lecture hall

Session 1

Chair: *Gernot Frenking (Marburg, Germany)*

08.30 – 08.35 Michael Springborg (Saarbrücken, Germany): *Opening*

08.35 – 09.20 I1: Dominik Marx (Bochum, Germany): *Recent trends in ab initio simulation*

09.20 – 09.40 C1: Daniel Sebastiani (Mainz, Germany): *Proton momentum-space densities from first-principles path-integral molecular dynamics simulations*

09.40 – 10.00 C2: Petr Slavíček (Prague, Czech Republic): *Photochemical reactions studied by ab initio quantum molecular dynamics: from molecules to clusters*

10.00 – 10.30 Break

Session 2

Chair: *Christoph van Wüllen (Berlin, Germany)*

10.30 – 11.15 I2: Richard L. Martin (Los Alamos, USA): *The localization / delocalization dilemma in the electronic structure of f-element oxides*

11.15 – 11.35 C3: Anatoly Titov (St. Petersburg, Russia): *Combined ab initio methods and accurate study of heavy-atom molecules to search for new physics beyond the standard model*

11.35 – 11.55 C4: Hélène Bolvin (Strasbourg, France): *Calculation of electronic g-matrices of actinide compounds*

11.55 – 13.30 Lunch break

Session 3

Chair: *W. H. Eugen Schwarz (Siegen, Germany)*

13.30 – 14.15 I3: John P. Perdew (New Orleans, USA): *Hyper-generalized gradient approximation for the exchange-correlation energy of density functional theory*

14.15 – 14.35 C5: Kartick Gupta (Bishnupur, India): *A DFT study of acidity of ortho- and para-substituted aromatic acids using molecular descriptors*

14.35 – 14.55 C6: Laszlo von Szentpaly (Stuttgart, Germany): *Critique and correction of conceptal DFT*

14.55 – 15.30 Break

Session 4

Chair: *Christel Marian (Düsseldorf, Germany)*

15.30 – 16.15 I4: Poul Jørgensen (Århus, Denmark): *A ground-state directed optimization scheme for the Kohn-Sham energy*

16.15 – 16.35 C7: Christian Ochsenfeld (Tübingen, Germany): *A linear-scaling AO-based MP2 method for large molecules by rigorous integral estimates*

16.35 – 16.55 C8: Andreas Köhn (Mainz, Germany): *Excitation energy transfer rates from correlated transition densities*

16.55 – 17.45 Break

Session 5

Chair: *Thomas Heine (Dresden, Germany)*

17.45 – 18.30 I5: Benoît Champagne (Namur, Belgium): *Ab initio polymer quantum theory: structural and vibrational properties*

18.30 – 19.30 Dinner

Poster Session 1

19.30 – 22.00 Posters with odd numbers. (Including free beverages)

Tuesday, September 18th

From 08.35 Registration in front of the lecture hall

Session 6

Chair: *Wolfgang Domcke (Munich, Germany)*

08.35 – 09.20 Presentation of the Hellmann Prize winner

09.20 – 09.40 C9: Serguei Fomine (Mexico City, Mexico): *Tubular aggregates of cyclic oligothiophenes. A theoretical study*

09.40 – 10.00 C10: Alisa Krishtal (Antwerp, Belgium): *The effect of structural parameters on the polarizabilities of methanol clusters: a Hirshfeld study*

10.00 – 10.30 Break

Session 7

Chair: *Gotthard Seifert (Dresden, Germany)*

10.30 – 11.15 I6: Benedetta Mennucci (Pisa, Italy): *Structure and properties of molecular solutes in electronic excited states: a quantum mechanical Polaizable Continuum Model*

11.15 – 11.35 C11: Jan-Ole Joswig (Dresden, Germany): *Proton transport in liquid phosphonic acid and polyphosphonic acid based polymers studied with molecular-dynamics simulations*

11.35 – 11.55 C12: Tillmann Klamroth (Potsdam, Germany): *Explicitly time-dependent configuration interaction simulations: dipole switching in large molecules*

11.55 – 13.30 Lunch break

Session 8

Chair: *Georg Jansen (Essen, Germany)*

13.30 – 14.15 I7: Joachim Sauer (Berlin, Germany): *Structure and reactivity of oxide catalysts*

14.15 – 14.35 C13: Judith Voll (Munich, Germany): *Laser induced femtosecond dynamics in the excited states of beta-carotene*

14.35 – 14.55 C14: Ozlem Ozcan (Düsseldorf, Germany): *A Monte Carlo – DFT study: adsorption of organosilanes on polar ZnO(0001) surfaces*

14.55 – 15.30 Break

Session 9

Chair: *Dirk Andrae (Bielefeld, Germany)*

15.30 – 16.15 I8: Arne Lüchow (Aachen, Germany): *Electron structure quantum Monte Carlo:recent developments and applications*

16.15 – 16.35 C15: Vincent Tognetti (Paris, France): *DFT study of ethylene dimerization by mixed bidentate P–N nickel complexes*

16.35 – 16.55 C16: Philipp Scherer (Munich, Germany): *Nonadiabatic coupling mechanism for ultrafast electron transfer in the reaction center of bacterial photosynthesis*

16.55 – 17.30 Break

Session X

Chair: *Wolfgang Domcke (Munich, Germany)*

17.30 – 18.00 X: Frank-Dieter Kuchta (DFG, Germany): *Die Deutsche Forschungsgemeinschaft informiert!*

18.00 – 18.30 Mitgliederversammlung AGTC

18.30 – 19.30 Dinner

Poster Session 2

19.30 – 22.00 Posters with even numbers. (Including free beverages)

Wednesday, September 19th

From 08.35	Registration in front of the lecture hall

Session 10

Chair:	*Daniel Sebastiani (Mainz, Germany)*
08.35 – 09.20	I9: Karlheinz Schwarz (Vienna, Austria): *DFT calculations of crystals and surfaces with WIEN2k*
09.20 – 09.40	C17: David Benoit (Ulm, Germany): *Accurate computation of vibrational spectra for extended systems*
09.40 – 10.00	C18: Liviu Hozoi (Dresden, Germany): *Correlated bands in oxides with wave-function based methods*
10.00 – 10.30	Break

Session 11

Chair:	*Beate Paulus (Dresden, Germany)*
10.30 – 11.15	I10: David Wales (Cambridge, UK): *Energy landscapes: from clusters to biomolecules*
11.15 – 11.35	C19: Konstantin Neyman (Barcelona, Spain): *Nanosized metal and oxide particles as realistic models of catalytic materials*
11.35 – 11.55	C20: Christoph Scheurer (Munich, Germany): *Simulation of photoluminescence spectra of neutral and charged exciton states in self-assembled semiconductor quantum dot molecules*
12.00 – 19.30	Excursion
19.30 – ?	Conference Dinner

Thursday, September 20th

From 08.35 Registration in front of the lecture hall

Session 12

Chair: *Frank Neese (Bonn, Germany)*

08.35 – 09.20 I11: Martin Kaupp (Würzburg, Germany): *Magnetic properties and local hybrid functionals*

09.20 – 09.40 C21: Dominik Kröner (Potsdam, Germany): *Laser-controlled chiral molecular switch: Quantum simulations for the stereoselective transformation between achiral and chiral atropisomers*

09.40 – 10.00 C22: Wolfgang Eisfeld (Bielefeld, Germany): *Taming of a beast: Multimode nonadiabatic excited state dynamics in* NO_3

10.00 – 10.30 Break

Session 13

Chair: *Michael Bühl (Mühlheim, Germany)*

10.30 – 11.15 I12: Hermann Stoll (Stuttgart, Germany): *Long-range correlation and local increments*

11.15 – 11.35 C23: Beate Paulus (Dresden, Germany): *Adsorption energy calculations with the method of increments: application to the adsorption of CO on the* CeO_2*(110) surface*

11.35 – 11.55 C24: Ulrich Wedig (Stuttgart, Germany): *Bonding properties in elemental Zn and Cd*

Closing

Chair: *Michael Springborg (Saarbrücken, Germany)*

11.55 – 12.15 Anne-Marie Kelterer (Graz, Austria): Presentation of the 44th Symposium for Theoretical Chemistry, 2008

Poster Prize Board: Presentation of the winners of the poster prizes

Closing

Lunch

Departure

Abstracts of invited lectures

Recent Trends in Ab Initio Simulation

Dominik Marx[1]

[1] *Lehrstuhl für Theoretische Chemie Ruhr-Universität Bochum 44780 Bochum; www.theochem.rub.de*

Ab initio simulation is a general class of approaches to computer simulation where the electronic structure problem is solved concurrent to the simulation. This can be achieved in molecular dynamics or Monte Carlo sampling frameworks, Car-Parrinello propagation in conjunction with density functional theory being a very efficient algorithm to do so which gives direct access to dynamical information of condensed phase systems.

This lecture will review recent developments in the field of ab initio simulation such as approximate quantum dynamics for computing the time evolution of anharmonic many-body systems and multideterminant propagation schemes for treating antiferromagnetically coupled spin systems. The former can be exploited to assess quantum effects on infrared spectra, whereas the latter allows one to address issues in molecular magnetodynamics. In addition, some representative applications will be discussed.

General References to ab initio simulation:

[1] J. Grotendorst, S. Blügel, and D. Marx (Editors), Computational Nanoscience: Do It Yourself! (NIC Jülich, 2006); see *www.theochem.rub.de/go/cprev.html*

[2] J. Grotendorst, D. Marx, and A. Muramatsu (Editors), Quantum Simulations of Complex Many-Body Systems: From Theory to Algorithms (NIC Jülich, 2002); see *www.theochem.rub.de/go/cprev.html*

The localization/delocalization dilemma in the electronic structure of f-element oxides

R.L. Martin[1]

[1] *Theoretical Division, Los Alamos National Laboratory, Los Alamos, NM*

The electronic structure of many of the oxides containing f-elements has long been a challenge for theory. For example, the traditional workhorses of density functional theory, the local density approximation (LDA) and the generalized gradient approximations (GGA), predict many of these systems to be metallic, when in fact they are insulators with band gaps of several eV. These problems reflect the localization/delocalization dilemma faced in systems with weak overlap and seem to be largely overcome by the new generation of *hybrid* density functionals developed for molecular studies. Only recently has it been possible to apply these functionals to solids, but in the cases studied thus far we find a distinct improvement. The hybrid functionals predict the correct insulating ground state, band gap, lattice constant and magnetic behavior at 0K, where

known. I will review the origin of the problem, how hybrid functionals differ from traditional ones, and recent applications to CeO_2 and Ce_2O_3, UO_2, and PuO_2, Pu_2O_3 and $PuO_{2.25}$.

Hyper-Generalized Gradient Approximation for the Exchange-Correlation Energy of Density Functional Theory

John P. Perdew,[1] Viktor N. Staroverov,[2] Jianmin Tao,[3] and Gustavo E. Scuseria[4]

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[2]*Department of Chemistry, University of Western Ontario, London, Ontario N6A 5B7 Canada*
[3]*Department of Physics, University of Missouri-Columbia, Columbia, MO 65211 USA*
[4]*Department of Chemistry, Rice University, Houston, TX 77005 USA*

In the density functional theory of Kohn and Sham 1965 [1], the ground-state electron density and energy can be found by solving one-electron selfconsistent Schroedinger equations. In practice, the density functional for the exchange-correlation energy must be approximated. Common approximations can be arranged on the rungs of a ladder according to the ingredients used to construct the energy density. The first rung or local spin density approximation [1] uses only the local spin densities. The second rung or generalized gradient approximation (GGA) [2] uses also the gradients of the spin densities. The third rung or meta-GGA [3] uses additionally the orbital kinetic energy densities for each spin. The addition of more ingredients permits the satisfaction of more exact constraints, with greater accuracy. The first three rungs have fully nonempirical constructions [1-3]. The fourth rung or hyper-GGA uses in addition the exact exchange energy density, and is able to satisfy many more exact constraints (full exact exchange, freedom from self-interaction error, etc.). However, it loses the automatic cancellation of error between approximate exchange and approximate correlation, and so requires the use of one or more empirical parameters. We present a new hyper-GGA that satisfies essentially all the exact constraints that a hyper-GGA can, and achieves high accuracy for atomization energies of molecules and energy barriers to chemical reactions.

[1] W. Kohn and L.J. Sham, Phys. Rev. **140**, A1133 (1965).
[2] J.P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. **77**, 3865 (1996).
[3] J. Tao, J.P. Perdew, V.N. Staroverov, and G.E. Scuseria, Phys. Rev. Lett. **91**, 146401 (2003).

A Ground-State Directed Optimization Scheme for the Kohn–Sham Energy

Stinne Høst,[1] Branislav Jansík,[1] Jeppe Olsen,[1] Poul Jørgensen,[1] Simen Reine,[2] and Trygve Helgaker[2]

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Kohn–Sham density-functional calculations are used in many branches of science to obtain information about the electronic structure of molecular systems and materials. The traditional method for optimizing the Kohn–Sham energy consists of a Roothaan-Hall (RH) diagonalization of the Kohn-Sham matix to obtain a new density matrix followed by an averaging of this density matrix and the density matrices from previous iterations to obtain an improved density matrix for the next iteration. The improved density matrix is usually found using the DIIS algorithm of Pulay. The RH/DIIS method suffers from fundamental problems that may lead to divergence or, even worse, convergence to an energy saddle point rather than to the ground-state minimum. This happens in particular, for the larger and more complicated electronic systems that are often studied by Kohn–Sham theory nowadays.

We here present a novel method for Kohn–Sham energy minimization that does not suffer from the flaws of the conventional approach, combining reliability and efficiency with linear complexity. In particular, the proposed method converges to a minimum, avoiding the sometimes spurious solutions of the traditional method and bypassing the need to examine the structure of the provided solution.

Ab Initio Polymer Quantum Theory: Structural and Vibrational Properties

Benoît Champagne[1]

[1] *Laboratoire de Chimie Théorique Appliquée, Facultés Universitaires Notre-Dame de la Paix, Rue de Bruxelles 61, B-5000 Namur (Belgium)*

Crystal orbital techniques based on periodic boundary conditions are a convenient way to describe the properties of stereoregular polymers [1]. Over the last 40 years, methods and algorithms have been elaborated at different levels of approximation, encompassing the reference Hartree-Fock level, Möller-Plesset perturbation theory, and more recently coupled-cluster approaches. This talk will review basic aspects of such approaches including lattice summations and long-range interactions, integration procedures, and quasi-linear dependencies. It will then concentrate on structural, vibrational, and mechanical properties and in particular it will discuss methods to compute the forces on nuclei, the Young elastic modulus, vibrational frequencies and intensities, as well as vibrational polarizabilities.

[1] B. Champagne, in Molecular Simulation Methods for Predicting Polymer Properties, edited by V. Galiatsatos (Wiley & Sons, New York, 2005), 1-46.

Structure and properties of molecular solutes in electronic excited states: a quantum mechanical Polarizable Continuum Model

Benedetta Mennucci [1]

[1] *Dipartimento di Chimica e Chimica Industriale, Uiversità di Pisa, vi Risorgimento 35, I-56126 Pisa, Italy*

The desciption of structures and properties of electronically excited states of molecular systems is a rather complex problem. In the years, the development of linear response approaches such as CIS, ZINDO and, more recently, TDDFT has contributed to make these studies possible also for realistic systems.

In parallel to this progress in the quantum mechanical description of the electronic states there has been a parallel extension of these studies to molecules embedded in different environments of various complexity, from standard solvents to interfaces between gas and solvent or between solvent and solvent, and to metal surfaces.

In this talk a review of the most recent advances in the field obtained by exploiting a polarizable continuum model [1] is presented and discussed.

[1] J. Tomasi, B. Mennucci, and R. Cammi Chem. Rev. **105**, 2999 (2005).

Structure and Reactivity of Oxide Catalysts

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The use of computational techniques based on quantum mechanics in concert with experimental techniques for understanding the structure and reactivity of complex solid catalysts is described. The discussion for metal oxide catalysts, in particular supported transition metal oxides [1,2] and zeolites [3], includes use of quantum mechanics and statistical mechanics to bridge the gap between ultra-high vacuum model systems (surface science) and catalysts under "real" conditions [4,5]; use of quantum mechanics and IR spectroscopy to analyse the differences and similarities of gas phase clusters and solid catalysts [1,6]; limits and merits of periodic models and (embedded) cluster models; the power of DFT and needs to go beyond DFT (multi-reference, dispersion); the use of hybrid techniques: QM:MM and QM:QM [3].

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Electron structure quantum Monte Carlo: recent developments and applications

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With Monte Carlo techniques the numerical quantum simulation of the full-dimensional Schrödinger equation is a viable and favorably scaling method that directly, without the expansion into orbitals, describes the electron correlation. Over the last few years, these quantum Monte Carlo (QMC) methods have shown to be applicable to a wide range of quantum chemical problems with high accuracy, including excited states and weak interactions. QMC can retain the inherent favorable scaling of Monte Carlo methods in these applications. Recent applications and improvements of QMC are discussed. An addition, new ways to control the fixed-node error of the diffusion quantum Monte Carlo method are presented.

Die Deutsche Forschungsgemeinschaft informiert!

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Das Ziel dieser Veranstaltung ist es, Ihnen aktuelle Informationen aus der Forschungsförderung der Deutschen Forschungsgemeinschaft zu vermitteln. Neben allgemeinen Hinweisen zu den koordinierten DFG-Programmen, insbesondere Schwerpunkten, Förderinstrumenten für Nachwuchswissenschaftler und internationale Kooperationsprojekten, stehen Themen im Mittelpunkt, die für ihr Fachgebiet, die Theoretische Chemie, von Interesse sind. Es ist zudem vorgesehen über die Antrags- und Bewilligungsentwicklung in der Einzelförderung, die Fachgutachterwahlen 2007, die Arbeit der Fachkollegien und das Begutachtungsverfahren zu berichten.

DFT calculations of crystals and surfaces with WIEN2k

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Density functional theory (DFT) made it possible to calculate the electronic structure of rather large systems. Various functionals (LDA, GGA, hybrid functionals, LDA+U) are used to provide a proper description of the electronic strucuture of different materials. The translational symmetry of a crystal allows to focus on a unit cell but surfaces can be treated as well using periodic boundary conditions and assuming a slab geometry with an atom-free region representing vacuum. Plane waves are a natural basis set for solids but they can not represent the oscillating wave functions near a nucleus. Therfore mixed basis schemes such as the linearized augmeted plane wave (LAPW) method provide a very accurate description for all elements in the periodic table. In LAPW the unit cell is partitioned into non-overlapping atomic spheres and an interstitial region, where plane waves are used. Inside each atomic sphere the plane waves are augmented by atomic-like functions with (numerically calculated) radial functions times spherical harmonics. In addition relativistic effects and the treatment of semi-core states can be included.

The WIEN2k computer package (see www.wien2k.at) is based on this method and is one of the most accurate code for solids (for a review see [1]). Presently it is used worldwide by more than 1100 groups in academia and industry. The concepts will be described and various applications will be given from minerals to magnetic materials or metal insulator transitions, or electric field gradients.

A recent application of WIEN2k is our study of a boron nitride surface layer that forms on a metal substrate [2]. When borazine is deposited on transition metal surfaces, in particular on Rh(111), an ordered structure, called nanomesh, forms that has a periodicity of about 3.2 nm. In order to simulate this surface structure a unit cell containing about 1100 atoms is used. Nowadays such calculations are feasible and the corresponding computer requirements will be sketched. This example will be used to illustrate the various possibilities of WIEN2k calculations using forces on the atoms, geometry optimizations, densities of states, workfunctions, spectra, etc.

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Energy Landscapes: from Clusters to Biomolecules

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Coarse-graining the potential energy surface into the basins of attraction of local minima, provides a computational framework for investigating structure, dynamics and thermodynamics in molecular science [1,2]. Steps between local minima form the basis for global optimisation via basin-hopping [3,4] and for calculating thermodynamic properties using the superposition approach [1,5] and basin-sampling [6]. To treat global dynamics we must include transition states of the potential energy surface, which link local minima via steepest-descent paths. We may then apply the discrete path sampling method, which provides access to rate constants for rare events [7-9]. In large systems the paths between minima with unrelated structures may involve hundreds of stationary points of the potential energy surface. New algorithms have been developed for both geometry optimisation [10] and making connections between distant local minima [11], which allow us to treat such systems. Kinetic analysis of large stationary point databases is facilitated by a new graph transformation approach [12].

Applications will be presented for a wide variety of atomic and molecular clusters, including systems bound by isotropic interatomic potentials such as the Lennard-Jones and Morse forms, clusters of dipolar spheres [13], and molecular clusters such as nanodroplets of water [14-16]. Results for folding, misfolding and aggregation of peptides and proteins illustrate how experimental time and length scales can now be addressed for such systems.

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Magnetic Properties and Local Hybrid Functionals

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Recent advances in development, validation, and application of DFT in the area of magnetic resonance parameters are reported, based on the ReSpect program system [1]. Spin-orbit effects have important consequences on NMR and EPR parameters. One example is the unusually large spin-orbit effects on Cu hyperfine couplings in blue-copper protein sites [2]. Higher-order spin-orbit effects may be included within a spin-polarized two-component framework [3]. The computation of NMR chemical shifts of paramagnetic systems has recently been extended to systems with arbitrary spin multiplicity, by incorporating the influence of zero-field splittings on the Boltzmann distribution of energy levels [4]. Shortcomings of the existing exchange-correlation functionals become particularly apparent when looking at the NMR/EPR parameters of transition metal systems. This has motivated us to develop new functionals within the "local hybrid" framework using position-dependent exact-exchange admixture [5]. The first results are very encouraging for both thermochemistry and kinetics. In addition, the first self-consistent implementation as "localized local hybrid potentials" has allowed the first calculations of NMR chemical shifts, with equally encouraging results [6].

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Long-range correlation and local increments

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DFT is nowadays the method of choice for large molecules and extended systems, but standard LDA, GGA, and meta-GGA functionals have problems for van der Waals interaction and do not allow for a systematic improvement.

Part of these weaknesses can be overcome by restricting DFT to a short-range interelectronic interaction and using wavefunction-based ab-initio methods for the complementary long-range terms. It is shown that a both accurate and efficient treatment is possible at intermediate values of the coupling constant [1,2].

Still, application to extended systems necessitates a local correlation treatment on the ab-initio side. One of the simplest devices is a many-body expansion in terms of correlation contributions from localized orbital groups. Such an incremental expansion, on top of HF calculations, has proven to yield reliable results, at moderate cost, for a variety of solids (van der Waals crystals, ionic systems, semiconductors, and even metals) [3,4]. A possible application of the incremental method within a mixed DFT / ab-initio scheme will be discussed.

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Abstracts of contributed lectures

Proton Momentum-space Densities from First-Principles Path-Integral Molecular Dynamics Simulations

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We present an approach to compute reciprocal space probability densities of light nuclei in complex condensed-phase systems, in particular the momentum density $n_{\mathrm{nuc}}(k) = |\Psi_{\mathrm{nuc}}(k)|^2$ of protons. This property reflects the quantum nature of the hydrogen wavefunction in terms of the spatial localization and delocalization. Naturally, the momentum density of a nucleus reacts to changes in its chemical environment, especially hydrogen bonding. Momentum-space densities $n_{\mathrm{nuc}}(k)$ can directly be measured via inelastic Neutron Compton Scattering techniques.

Our method is based on finite-temperature path-integral Car-Parrinello molecular dynamics simulations. In order to obtain momentum-space densities, a modified version of the path-integral technique is used, which relies on linear ("open") paths for the particles of interest. The method is benchmarked on test systems and small molecules, and further applied to describe the ferroelectric and paraelectric states in potassium dihydrogen phosphate (KDP) at different temperatures. In such systems, the momentum distributions $n_{\mathrm{nuc}}(k)$ allow for a distinction between temperature-induced hopping and quantum tunneling states of the hydrogen-bonded protons.

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Photochemical reactions studied by *ab initio* quantum molecular dynamics: From molecules to clusters

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In the *ab initio* molecular dynamics approach, the equations of motion for both electrons and nuclei are solved simultaneously as the simulation proceeds (direct dynamics). This approach represents a hopeful tool for revealing reaction mechanisms: the dynamical equations are automatically leading us to important structures without any bias or prejudice while bond breaking or bond making processes are properly described. In many important cases the nuclear equations of motion have to include quantum effects. Specifically, quantum (or at least semiclassical) dynamics has to be employed for a description of non-adiabatic processes or when tunneling of light particles is involved. This is almost always the case in photochemistry. Full Multiple Spawning (FMS) method [1] is one route for this connection between quantum molecular dynamics and electronic structure calculations. Methods eof extedning the scope of this approach beyond simple molecules will be discussed [2].

We will show how a combination of quantum chemical and ab initio quantum dynamical calculations can be used for an exploration of photochemical reactions involving non-adiabatic processes. Two molecular examples will be provided: photochemistry of glycine [3] and photochemistry of nitrophenols.

Many important photochemical processes occur in the bulk phase or on the surface of nanosized particles. Information obtained by a study of isolated molecules can thus be of a limited relevance. Recently, we have started to study photochemistry of molecules assembled in molecular cluster in a close collaboration with an experimental group. Two examples will be shown: study of photochemistry of pyrrole molecular clusters [4] and study of hydrogen halide acidic and photochemical dissociation on the surface of water clusters [5].

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Combined *ab initio* methods and accurate study of heavy-atom molecules to search for new physics beyond the Standart model

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The search for the space (P) and time-reversal (T) parity nonconservation effects is of fundamental importance for modern theory of fundamental interactions and symmetries and immense experimental effort is invested in the search for the fingerprints of P,T-parity nonconservation effects, such as the electron electric dipole moment (eEDM) [1]. The tightest bound on the value of eEDM, d_e, was obtained in the experiment on Tl atom, which established an upper bound of $|d_e| < 1.6 \times 10^{-27} e\cdot$cm ($e$ is the charge of the electron). Molecular systems provide a way for much enhanced sensitivity since the effective intra-molecular electric field acting on electrons in polar molecules can be five or more orders of magnitude higher than the maximal field available in a laboratory. Such experiments were successfully performed so far only on the molecular YbF beam. Recently some new approaches were suggested, such as the search for P,T-odd effects in molecular cations PbO* [2], HI^+, HfF^+ etc. [3]. The new generation of eEDM experiments will have a sensitivity of $10^{-30} \div 10^{-28} e\cdot$cm. Their results are expected to dramatically influence all the popular extensions of the Standard model, in particular supersymmetry, even if bounds on the P,T-odd effects compatible with zero are obtained [1]. For preparation and interpretation of these experiments, accurate calculations of electronic densities on the heavy nuclei are required and different molecular properties should be computed.

Precise calculations of core properties in heavy-atom systems, like to hyperfine constants and P,T-parity nonconservation effects, which are described by the operators heavily concentrated in atomic cores, usually require accurate accounting for relativistic and correlation effects. However, precise all-electron four-component treatment of molecules and clusters with heavy elements is yet rather consuming. In turn, the relativistic effective core potential (RECP) calculations of valence (spectroscopic, chemical etc.) properties of such molecules are very efficient because the two-component RECP method allows one to reduce dramatically the computational cost providing a good accuracy of correlation and relativistic treatment for the valence electrons. However, the valence molecular spinors are usually smoothed in atomic cores and, as a result, direct calculation of electronic densities near heavy nuclei is impossible. The former circumstance stimulated further development of the RECP approaches and, in particular, the generalized RECP concept was developed [2] exploiting the idea of different treatment of inner core, outer core and valence shells. The latter had led to the methods of *nonvariational and variational one-center restoration* of correct shapes for four-component spinors in atomic cores after a two-component RECP calculation of a molecule [2]. In the report, the RECP and restoration methods are discussed. Their efficiency is illustrated in benchmark to-date correlation calculations of hyperfine structure and P,T-parity nonconservation effects in polar molecules YbF, PbO [2], HfF^+ [3], and liquid Xe [4] etc., which are of particular interest for the experiments on search for a new physics beyond the Standard Model.

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Calculation of electronic g-matrices of actinide compounds

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The molecular g-factors are crucial parameters in Electron Paramagnetic Resonance (EPR) spectroscopy. They parametrize the Zeeman effect that involves the interaction of a spin magnetic moment with a magnetic field. Before the 90's, g-factors were mostly calculated by semi-empirical methods while in the last decade, ab initio techniques have been developed based on wave functions or density functional theories either using a sum over states (SOS) expansion or using response theory [1]. We have recently proposed an alternative method based on wave function theory including spin-orbit effects by the SO-RASSI method [2]. This method has been applied with success to small molecules containing atoms until the third series of the transition metals and to a mixed-valence bimetallic complex [3] and the aim of this work is to applied it to actinide complexes. To our knowledge, only few attempts have been made to calculate g-factors of actinide complexes from first principles [4,5]. Relativistic effects in actinide complexes are important, so the deviation of g-factors from the value of the free electron is large and g-factors can even become negative. Most of the experiments measuring magnetic properties of actinide complexes date from the sixties. EPR and magnetic susceptibilities give the absolute values of the g-factors while experiment using cicularly polarized light provide the sign of the product of the three factors. Magnetic properties of these compounds are easily understood using ligand field theory models.

In this presentation, we first point out the arbitrary character of the g-matrix and specially discuss its transformation under a rotation in the spin space and we show that only the G tensor $gg^\dagger$ has a physical significance. Calculations first concern complexes with $5f^1$ configuration like the AnX_6^{q-}series with An = Pa,U,Np and X = F,Cl,Br and the NpO_2^{2+} cation either in the gas phase or with equatorial ligands. For the latter, the influence of the nature on magnetic properties of the equatorial ligands is discussed. Finally, the magnetic properties of Np^{4+} in $\mathrm{Cs_2ZrCl_6}$ crystal are calculated. The ground state is in this case orbitally degenerate and cubic terms have to be added to the spin Hamiltonian. Both the linear term, namely the g-factor, and cubic terms are deduced from our calculations and compared to experimental values.

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A DFT Study of Acidity of Ortho- and Para-Substituted Aromatic Acids Using Molecular Descriptors

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Quantum chemical computations with full geometry optimization at the B3LYP/6-31G(d) level in aqueous phase at 298K are performed to calculate a set of molecular properties for 53 aromatic organic acids like benzoic acid, phenol, phenylacetic acid, cinnnamic acid, benzohydroxamic acid, benzenesulfonic acid, anilinium ion and their meta- and para-substituted derivatives. Molecular descriptors like ionization potential, electron affinity, hardness, chemical potential, global electrophilicity and free energy of deprotonation of these acids are computed. The local descriptors like Fukui functions and group philicity (Wg+) are calculated with MPA and HPA schemes. These aromatic acids are reacted with a strong base, OH-. The computed Gibbs free energy of deprotonation, the fractional numbers of electrons transferred,DeltaN and electrophilicity based charge transfer index, electrophilicity based charge transfer index (ECT) in acid-base reaction with a strong base, OH- and Wg+ are correlated with the experimental pKa values of these acids. It is found that this approach is particularly effective in discussing trends of changes in acidity of intimately related molecules. The effect of substituent on these descriptors is also studied. These parameters are correlated with experimental Hammett substituent constants (sigma). ECT and fractional number of electron transfer (DelN) correlate strongly with pKa and ? in separate groups of aromatic acids. Minimum energy, maximum hardness and minimum electrophilicity principles are tested for the acid-base reactions.

Key Words: Aromatic acids, Gibbs free energy, Hammett substituent constant, electron transfer, Electrophilicity based charge transfer index.

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Critique and Correction of Conceptual DFT

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Conceptual DFT (CDFT) and its applications have been the object of about one thousand research papers [1, 2]. CDFT assumes that the energy, E, is a continuous and differentiable function, E(N), of the number of electrons, N. The chemical potential, μ, and the chemical hardness, η, are defined as the first, and second partial derivatives of E(N) with respect to N. The chemical potentials of two systems, X and Y, are equalized as the systems start interacting. The CDFT electrophilicity index, ω, has found wide application [3-5].

A closer scrutiny of the foundations and applications of CDFT reveals, however, that it leads to severe contradictions to chemical principles and reality:

(i) The variation principle is violated; the CDFT equalization of μ lowers the energy of separated systems at large distances much below that of the isolated systems.

(ii) The charge transfer between X and Y is too small by a factor two. For the classical case of one-electron transfer at no cost, i. e., I(X)=A(Y) the equalization of μ is achieved by transfering just 0.50 electronic charge. Even for ionic diatoms, e. g., CsCl, the CDFT partial charge is below 0.50. Thus CDFT dipole moments are systematically too small.

(iii) According to the CDFT electrophilicity index the maximum uptake of electrons by any atom or molecule is smaller than one [3, 4], unless $A \geq I/3$. Stable anions are practically not existing according to CDFT.

The problems and contradictions are traced back to the occurance of spurious electron-electron attraction in case of fractional charges on the subsystems, X and Y [4, 6, 7]. Recent progress in properly incorporating the chemical context [7, 8] is reported. A different parabolic function E(N) for a defined chemical context, i. e., bond formation, accounts for the shared-pair aspects in charge transfer processes within chemical bonding. All of the above contradictions are resolved in this context, and very sensible results are obtained [4, 6, 7]. A generalized Born-Haber cycle with a common asymptotic reference energy for less polar solids and diatomic molecules is presented [7].

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A Linear-Scaling AO-Based MP2 Method for Large Molecules by Rigorous Integral Estimates

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Describing electron correlation effects for large molecules is a major challenge for quantum chemistry due to the strong increase of the computational effort with molecular size. In order to overcome this limitation, we present a rigorous method based on an AO-formulation of MP2 theory [1], which allows to avoid the conventional fifth-power scaling of MO-MP2 theory and to reduce the scaling to linear without loss of accuracy. The key feature of our method are multipole-based integral estimates (MBIE) [2,3], which account for the 1/R coupling in two-electron integrals and allow to rigorously preselect integral products in AO-MP2 theory. Here, the magnitude of products decays at least with $\frac{1}{R^4}$, so that a linear-scaling behavior can be achieved by numerical thresholding without sacrificing accuracy. The linear-scaling increase of the computational effort is reached much earlier than for HF or DFT approaches: e.g. the exact behavior of products indicates a scaling of $N^{1.0}$ from one to two DNA base-pairs for a 6-31G* basis. The number of significant elements in the pseudo-density matrices and of shell pairs hints to a very similar linear-scaling behavior for larger basis sets studied up to cc-pVQZ. First results of a preliminary implementation [4] show that an early crossover to conventional MP2 schemes for systems smaller than two DNA base pairs is observed, while already for a system with four DNA base pairs wins are at least a factor of 16 and the same accuracy is provided. Due to the linear-scaling behavior, these wins increase dramatically with system size.

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Excitation energy transfer rates from correlated transition densities

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Excitation energy transfer (EET) takes place if an electronically excited donor molecule decays to its ground state by simultaneous excitation of a neighbouring acceptor molecule. The process is of ultimate importance for the photophysical properties of e.g. biochemical systems or polymers and the distinct $1/R^6$ distance dependence of the leading dipole-dipole contribution to the EET rate (Förster Theory [1]) is widely applied for distance measurements on the nanometer scale.

One approach to the calculation of the electronic contributions to EET coupling originates from symmetry-adapted perturbation theory and requires the knowledge of the transition densities of the monomers only. Owing to the size of the involved molecules, the vast majority of such investigations were carried out with semi-empirical models [2], however, more recently, transition densities from time-dependent density functional theory (TDDFT) have been employed as well [3].

Here, we report an implementation of the EET coupling matrix elements which accepts one-particle transition densities from arbitrary methods, ranging from configuration interaction with singly excited determinants (CIS) and the random phase approximation (RPA) to coupled-cluster theory. Particular interest is paid to a special coupled-cluster model, RI-CC2 [4], which is well suited to the treatment of fairly large chromophores.

For coupled-cluster methods, however, some surprising inconsistencies arise when comparing EET rates from the perturbational approach to those obtained from supermolecular calculations. These problems can be traced back to an incorrect separability of the two-particle transition density matrix which will be discussed in some detail. For the long-range interaction regime, it will be argued that results from the perturbational approach are superior to supermolecular calculations.

As an application, we discuss the EET rates in rylene-dyads, which have been previously studied by single molecule spectroscopy [5]. The questions in focus are (a) the role of bridge-assisted energy transfer and (b) the validity of Förster-theory for dyes with extended chromophores.

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Tubular aggregates of cyclic oligothiophenes. A theoretical Study

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Creation of hollow tubular structures by noncovalent self- assembly of appropriately crafted organic molecules has been the subject of considerable research in recent years [1]. Well-defined pi-conjugated macrocycles are of interest as modular building blocks for the assembly of new materials [2] and supramolecular architectures. Due to their toroidal structure, they could represent molecular circuits which would additionally include sites for recognition and selective complexation. In this respect, it is necessary to mention recently synthesized fully conjugated macrocyclic structures: cyclo(oligothiophene-diacetylenes)and cyclo[n]thiophenes [3]. capable of self-assembling. We have recently reported a theoretical study of oligothiophene catenanes and knots in different oxidation states [4] showing that the electronic properties of conjugated molecules depends in great extent on supramolecular architecture. The goal of this contribution is to study the effect of tubular aggregations of cyclic oligothiophenes on their electronic properties. The geometries of neutral, mono- and diionized cyclo[8]thiophene and the corresponding tubular aggregates containing 2,3,4 and 5 cyclo[8]thiophene units were fully optimized at MPWB1K level of theory using 3-21G* basis set. Closed and opened shell systems were calculated using RHF and UHF methods, respectively. Recently defined MPWB1K functional performs very well for non-bonding interactions which are of primary importance for tubular assembling of cyclooligothiophenes. Additional calculations carried out on cyclo[4]thiophene dimer showed that MPWB1K and MP2 models gave very similar geometries, while other popular functionals such as B3LYP and PBE0 strongly underestimate pi-pi stacking energies between thiophene fragments predicting significantly larger distances between cyclo[4]thiophene units in dimer compared to MP2 and MPWB1K models. It was found that binding energies between cyclo[8]thiophene macrocycles vary from 44.6 kcal/mol for dimer to 42.7 kcal/mol for pentamer in case of neutral aggregates. In case of monoionized species the binding energy increases (50.8 for dimer and 47.0 kcal/mol for pentamer) reflecting additional stabilization of positive charge by neighbour macrocycles. On the other hand, in case of dications the binding energy drops to 11.1 kcal/mol for dimer, further increasing to 42.8 kcal/mol for pentamer due to electrostatic repulsion. Calculations show that the most stable dicationic state for tubular aggregates is open-shell singlet. The energy difference between triplet and singlet states decreases with the number of units in tubular aggregates reflecting dissociation of a bipolaron into two separated polarons. This conclusion is also confirmed by the charge distribution analysis for charged tubular aggregates; In case of pentamer cation-radical the most of the positive charge is located at the middle macrocyle, while in case of dications the first and the last macrocycles are the most positively charged ones. The band gap estimation carried out at TD-B3LYP/6-31G* level reveals that even for pentamer (2.49 eV) it close to that found for isolated monomer (2.60 eV) rendering them as semiconductors. In conclusion tubular aggregates of cyclic oligothiophenes are stable formations with significant binding energy between macrocycles. They can be oxidized similar to linear polythiophene producing stable mono and dicationic species rendering them as attractive synthetic target for supramolecular chemistry.

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The Effect of Structural Parameters on the Polarizabilities of Methanol Clusters: A Hirshfeld Study

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Solvents form an interesting and challenging topic, which has been widely studied in the literature in the past decades. Although much progress has been already made in this field, many questions still remain in particular concerning the effect of solvents on the structures and properties of solutes. In order to try and understand those effects, one can start by investigating the structure and properties of the pure liquids, where single molecules can be regarded as a solute, surrounded by identical solvent molecules. In this study the dynamic structure of a liquid is approximated by the structure of clusters.

Although the structure of a single molecule in a cluster is simple to obtain, the knowledge of its properties usualy requires a partitioning method, of which the Hirshfeld method [1] is our method of choice. The Hirshfeld method was introduced in 1977 primarily as a method for the partitioning of charge density. Later it was extended for the partitioning of properties such as dipole moments, quadrupole moments, polarizabilities, energies and and others. Recently, Bultinck et. al. [2] have revised this method and proposed an iterative version, Hirshfeld-I, which is more in line with information theory.

The Hirshfeld-I method was used in this study for the analysis of polarizabilities of methanol clusters. The choice of the polarizability as the first property to be investigated is made because of its importance in the solvation process. The polarizabilities of fifty different methanol clusters, with aggregation numbers ranging between 2 and 12, were investigated. In a previous study on water clusters [3], the polarizabilities of water molecules in water clusters were found to be strongly and straightforwardly dependend upon the hydrogen bond network. The introduction of a methyl group into the structure of the molecule creates a more complicated system. Whereas the water molecule is a symmetric entity, which can form up to four hydrogen bonds when placed in a cluster (DDAA), the methanol molecule can only form three hydrogen bonds (DAA), due to the replacement of one hydrophilic part (OH) by an hydrophobic part (methyl group). Whereas the polarizabilities of the hydroxyl groups follow the trends established in the water clusters, the polarizabilities of the methyl groups exhibit a strongly different behaviour. The hydrogen bonding network as well as the polarizabilities are stronly affected by structural and sterical factors. The methyl groups were found to be especially sensitive to these effects. Finally, the influence of BSSE was studied by means of the counterpoise method and was found to be significant, increasing the polarizabilities of the water molecules by upto 10 percent.

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Proton transport in liquid phosphonic acid and polyphosphonic acid based polymers studied with molecular-dynamics simulations

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Polyphosphonic acids are promising candidates as proton conductors in proton-exchange membrane fuel cells. However, the proton conduction mechanism in these systems is only poorly understood. Atomistic molecular-dynamics (MD) simulations can considerably contribute to the understanding of the conduction process and the transfer mechanisms [1].

Currently, we are focussing on the proton transfer in liquid phosphonic acid and in model polymer structures. Therefore, we utilize density-functional based MD simulations [2]. For the liquid phosphonic acid we analyze the speed of the proton transfer (hopping rates), pair-distribution functions, diffusion coefficients, and geometrical conditions for the transfer. The influence of an excess charge carrier is also studied.

Using the results obtained from the simulations of the liquid, we simulate model polymer structures, e.g. poly-vinyl phosphonic acid. Here we can study the proton transfers along and between polymer chains varying their distance and relative arrangement. The role of additional water molecules is studied in both cases.

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Explicitly time-dependent configuration interaction simulations: Dipole switching in large molecules

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We report laser-driven charge transfer simulations for Li-(Ph)$_n$-CN ($n = 1, 2, 3$) using the TD-CIS (Time-Dependent Configuration Interaction Singles)[1] approach. These molecules are used as systematically extendable model systems, in order to investigate the selectivity, and thus controllability, of an ultra-short laser-induced electronic excitation as a function of the molecular size. For example, such control may be needed if a small electronic molecular switch is embedded in a larger molecular device.

We employ ultra-short π-pulses in order to induce an electronic excitation, *i.e.* a charge transfer transition, using the TD-CIS method within the fixed nuclei approximation[2,3]. The focus of our dynamical calculations lies on the selectivity of these π-pulses depending on their temporal width, and the molecular size. We show that for pulse widths larger than 1000 $\hbar/E_h$ the selectivity is lost mainly due to multi-photon transitions and dynamic polarizations of the molecules. For the latter processes a systematic dependence on the molecular size is observed, *i.e.* they get more important for larger molecules. Therefore, we believe that special care should be taken when simulating electronic transitions in extended molecular systems to include effects due to linear and non-linear polarizabilities. For large molecules and/or short and intense laser pulses few state models might actually overestimate the selectivity of laser excitations.

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Laser induced femtosecond dynamics in the excited states of β-carotene

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We present a three dimensional three state model for the theoretical description of the laser induced quantum dynamics, based on experimental, quantum-chemical and -dynamical data of β-carotene. From the electronic ground state the system is excited by a femtosecond pulse into the optically allowed S_2 state. Internal conversion to the vibrationally hot S_1 state via conical intersections is performed in about 180 fs. Here during 700 fs the system cooles down. The three state model was fitted to data of control experiments performed on β-carotene in the group of M. Motzkus. Here the internal conversion was followed by transient detection of the Raman active C-C and C=C stretch modes. Quantum chemical ab initio calculations were performed to specify the kinetic Hamiltonian, the Raman modes involved, their force constants and the relative position of the interacting potentials. With this model the mechanism of internal conversion is studied, incoorporating vibrational cooling in the S_1 state. The results are used to investigate the controllability of the transfer rate by modulated laser pulses.

A Monte Carlo - DFT study: Adsorption of organosilanes on polar ZnO (0001) surfaces

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The ultimate aim of the project is to investigate the mechanism of organosilane adsorption on zinc oxide surfaces using computational tools and various experimental techniques. The study of the processes during adsorption requires the selection of a proper model system to describe single molecule - surface interactions and to reveal the binding mechanisms.

In this study, polar ZnO (0001) surface is selected as the model substrate due to its abundance in amorphous zinc oxide. The main challenge in modeling of ZnO (0001) surfaces is the determination of the stabilization mechanisms to cancel the dipole moment between Zn and O terminated surfaces. Surface reconstructions, adsorption of OH^- on the Zn terminated surface and use of pseudo-oxygen atoms with 6.5 valency at the O terminated surface have been tested using SIESTA (Spanish Initiative for Electronic Simulations with Thousands of Atoms) code. The binding energies of a variety of molecules to clean, hydroxylated and reconstructed surfaces have been computed to determine the most favorable binding state.

For the verification of computed binding energies, a Monte Carlo program[1,2] is developed that uses the binding energies calculated with SIESTA as input and produces the Thermo-Desorption

Spectra. The comparison of different ZnO (0001) surface stabilization mechanisms, and the results of TPD experiments along with the Monte Carlo analysis will be discussed in the presentation.

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DFT study of ethylene dimerization by mixed bidentate P-N nickel complexes

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Short chain length α-olefins are strategic targets in industrial chemistry. Among the nickel(II) catalysts for ethylene dimerisation [1], the mixed bidentate P-N nickel complexes present some remarkable advantages [2]. In particular, the simplicity of their synthesis [3] and their excellent activity constitute strong arguments to deepen the explanation of the dimerisation process. Therefore a DFT study of the mechanism seems a crucial step to achieve a more precise understanding of the reaction process and to develop new catalytic systems.

Two mechanisms are investigated for three asymmetric model systems with various hindrances. The first one is the Cossee mechanism, previously studied for symmetric diimine nickel complexes by Musaev and al. [3]. The second one is the metallacycle mechanism that was proposed thirty years ago by Grubbs for monodentate nickel(0) complexes [4].

It was found that butene is always predominant for both mechanisms and that the elimination barrier is almost independent of the substituents on the nitrogen atom. On the contrary, the whole insertion process is all the more disfavoured as hindrance increases. The third insertion is actually the sum of two steps: the insertion itself and also the previous coordination of ethylene. In the hypothesis of a Cossee mechanism, this coordination occurs on the most destabilizing side (in trans of the phosphorus atom). This trans effect was linked with the increase of the Ni-N bond length. Moreover, the evolution of the NPPC dihedral angle suggests that the selectivity is all the best as the plane conformation of the ligand is more stabilized.

Finally, none definitive argument has been found to distinguish between both mechanisms, but several ways of improving the catalysts efficiency are deduced from the calculations.

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Nonadiabatic Coupling Mechanism for Ultrafast Electron Transfer in the Reaction Center of Bacterial Photosynthesis

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We investigate ultrafast electron transfer as a transition between adiabatic Born-Oppenheimer states which include the mixing of donor and acceptor orbitals

$$\phi_i = (c\phi_D + s\phi_A) \rightarrow \phi_f = (-s\phi_D + c\phi_A)$$

where the mixing coefficients depend on the energy gap ΔE and the resonance integral J .

$$cs = \frac{J}{\sqrt{\Delta E^2 + 4J^2}}$$

Within the harmonic parallel mode approximation the nonadiabatic matrix element for a transition involving creation of one quantum of the promoting mode s reads

$$V_{nad,s} = FCF \frac{1}{2Q_{s,0}} \int \phi_i(r;R) \bigtriangledown_{Q_s} \phi_f(r;R) dr$$

The Franck-Condon factor of the accepting modes was calculated for model systems of Bacteriochlorophyll and Bacteriopheophytine molecules on the basis of 6-31G/HF calculations. The electronic factor was evaluated with the help of the commutator relation $[\bigtriangledown_R, H] = \Delta E \partial V_{pot}/\partial R$ as

$$V_{nad,s} = FCF \quad \int \phi_i(r;R) \left(Q_{s,0} \bigtriangledown_{Q_s} V_{pot}\right) \phi_f(r;R) dr$$

and the potential energy was approximated by the Coulombic interaction with atomic point charges. Within these approximations the electronic factor was evaluated for the model systems on the 6-31G/HF basis by calculating all necessary two- and three center integrals. We found an almost continuous distribution of accepting and promoting modes. Our estimate of the transfer rate on the basis of the nonadiabatic coupling is much larger than the estimate by the conventional Marcus expression for the same values of the resonance integral J and the Franck-Condon factor giving time constants in the picosecond region even for resonance integrals as small as $10cm^{-1}$. The nonadiabatic coupling can be visualized as a dipole-dipole type interaction where the infrared dipole of the promoting mode is coupled to the transition dipole of the electronic transition, which is essentially given by cs times the change of the permanent dipole moment. A similar treatment was recently presented for ultrafast internal conversion processes during electron solvation [1].

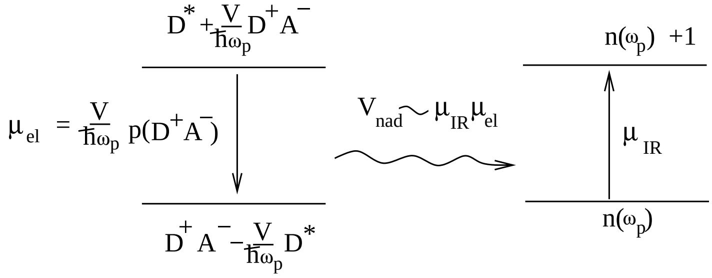

figure1: nonadiabatic dipole-dipole coupling mechanism

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Accurate computation of vibrational spectra for extended systems

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The prediction of accurate vibrational spectra from *ab initio* data has always been a challenging task in computational chemistry. One of the most promising methodologies to date that can simulate the vibrations of molecular systems of more than a handful of atoms is the vibrational self-consistent field technique and its extensions, combined with powerful parallelised quantum chemistry programs. This type of approach is very accurate, but the advantage of accuracy is often negated by the heavy demand on computational time. For this reason, the harmonic approximation has so far prevailed for most computational simulations of experimental vibrational data of large systems. In order to bring larger molecules within the reach of the self-consistent field techniques, we have developed over the past few years a new methodology for performing fast correlation-corrected vibrational self-consistent field (CC-VSCF or VCI) calculations using *ab initio* potential energy points generated on demand. This new method is based on an efficient reduction in the number of normal modes pair-coupling elements used in the CC-VSCF and VCI procedure. This approach saves computational time by focussing on the dynamically relevant part of the potential energy surface and neglecting the unused parts, leading to significant speedups compared to the usual VSCF approach, without significant loss of accuracy [1,2]. I will present our latest methodological developments and show applications of this new technique to achieve large speed improvements over the usual *ab initio* post-VSCF calculations

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Correlated bands in oxides with wave-function based methods

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We apply wave-function based quantum chemical methods to the study of electron correlation effects on the band structure of oxide compounds. Our analysis is based on a local Hamiltonian approach and performed on finite fragments cut from the infinite solid. As a first example we choose MgO, a relatively simple, weakly correlated ionic system. Localized Wannier functions and embedding potentials are obtained from prior periodic Hartree-Fock (HF) calculations. We investigate the role of various correlation effects in reducing the HF gap and modifying the band widths. On-site and nearest-neighbor charge relaxation as well as long-range polarization effects are calculated. Whereas correlations are essential for computing accurate band gaps, we found that in MgO they produce relatively small changes on the HF band widths. Our results [1] are in good agreement with the data derived from thermoreflectance and photoemission measurements.

As a second example, we discuss the electronic structure of the copper oxide superconductors. We focus on the role of the underlying antiferromagnetic lattice in renormalizing the effective quasiparticle hoppings. Such effects are not accessible by standard density-functional calculations. In particular, the nearest-neighbor effective hoppings are reduced by a factor of 4 [2]. We show that the dispersion of the lowest bands observed in photoemission is reproduced with quantitative accuracy [2,3]. Additionally, the evolution of the Fermi surface with doping, from small hole pockets in the deeply underdoped region to a holelike shape with a pseudogap at intermediate dopings and an electronlike appearance in the overdoped regime, follows directly from our *ab initio* data. Our results thus open a new avenue for the first-principles investigation of the electronic structure of strongly correlated transition-metal oxides.

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Nanosized Metal and Oxide Particles as Realistic Models of Catalytic Materials

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It is now well recognized that the difference in complexity between single crystal surfaces and "real" catalysts cannot be easily bridged. This is one of the major obstacles, known as material gap in heterogeneous catalysis, which notably hinders the extension of profoundly understood processes on single crystal surfaces to equally good understanding of heterogeneous catalysis. Indeed, active components of working heterogeneous catalysts are commonly present in the form of rather large nanoparticles, exposing a variety of point and extended defects that in many cases determine the reactivity. Thus, for realistic modeling of such systems one should go beyond the single crystal description, which, however, continues to be very useful reference. To this end, so- called model catalysts in the form of well-characterized metal nanoparticles supported on ordered oxide films have been proven experimentally very fruitful [1]. The presentation describes our modeling strategy [2], developed to computationally represent at a sufficiently realistic level oxide-supported nanoparticles with 10**3 metal atoms, which are experimentally explored as model catalysts. This novel approach employs ordered three- dimensional nanoparticles of 10**2 atoms with structures cut from bulk and terminated by low-index surfaces; in this size range cluster properties are scalable to the bulk. Our density functional calculations of clusters from

Pd55 to Pd146 showed that starting from 80 atomic particles the adsorption properties no longer exhibit size effects [2]. Thus, these models enable a realistic description of adsorption properties and reactivity of notably larger metal particles of similar shape in supported model catalysts [3]. Later applications of this modeling approach deal with structure of bimetallic PdZn particles containing 140 atoms and interactions with them [4], surface [5] and subsurface [6] impurities of light element atoms as well as reactions of methoxide (CH3O) decomposition on Pd nanoparticles [7]. Very recently, formation of arrays (nanowires) from Pd nanoparticles by self-assembling has been investigated [8] and extension of the modeling approach to metal-oxide particles of 200 atoms or more has been performed [9]. Examples of the above mentioned density functional applications of the nanoparticle modeling approach will be presented.

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Simulation of Photoluminescence Spectra of Neutral and Charged Exciton States in Self–Assembled Semi–Conductor Quantum Dot Molecules

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Semiconductor quantum dots (QDs) have attracted much interest since Qubits formed from interband charge excitations (excitons) represent a particularly promising system for quantum computation that can be coherently manipulated using picosecond laser pulses. An important step is to achieve coherent coupling of quantum states in spatially separated QDs to realize a system with conditional coherent dynamics that can be electrically and optically manipulated.

In recent experiments on vertically stacked self assembled InGaAs/GaAs QD–molecules (QDMs) control of the coherent coupling between excitonic states in the upper and lower dots was achieved by tuning the electric field oriented along the axis of the QDM. Single molecule spectroscopy revealed a clear anticrossing of spatially direct (e, h in the upper dot) and indirect (e, h in different dots) single exciton states. Good accord was obtained with realistic calculations of the single exciton spectrum of the QDM, confirming that the tunnel coupling is mediated principally by the electron component of the exciton wavefunction.[1]

More recently the influence of electrostatically introducing an additional electron into the QDM was studied. This leads to the formation of negatively charged Trions for which the spectrum of anti–crossings reflects the complex interplay between Coulomb couplings and the electrostatic energies of the two dots. Trion states in QDMs can be either spatially direct (e.g. $2e + 1h$ in upper dot) or indirect (e.g. $1e + 1h$ in upper dot and $1e$ in lower dot) and can be tuned into resonance by tuning the static electric fields. Furthermore, radiative decay takes place to a $1e$ final state that, itself, can hybridize over the two dots. Each anticrossing ($1e+1h$, $2e+1h$ initial and $1e$ final states) occurs at a distinct field, allowing for their clear identification. The richness of the spectra is further enhanced by spin–spin effects whereby singlet and triplet Trion states exhibit remarkably different behavior.

A simple model describing all possible spin–adapted many–particle states in the effective mass approximation yields a detailed analysis of the spectra. The excellent agreement with the experiment allows us to assign all observed features with confidence.[2]

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Laser-controlled chiral molecular switch: Quantum simulations for the stereoselective transformation between achiral and chiral atropisomers

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The influence of molecular chirality on the outcome of chemical or biochemical processes is still an important topic of today's research. For instance, reaching perfect control of the stereoselectivity in a chemical reaction remains one of the great challenges in chemistry. Traditional chemistry offers a variety of effective and well-established methods for stereoselective synthesis, e.g. by employing a chiral catalyst which drives the reaction towards the desired stereoisomer. In this case, a change of the handedness of the catalyst, e.g. induced by an external laser field, controls which stereoisomer is produced by the reaction. Indeed, chemical compounds, so-called chiroptical switches, exists that undergo isomerization upon irradiation at the appropriate wavelength resulting in a change of the chirality of the system [1]. In addition, various applications of chiroptical switches are possible in the field of nanoscale devices ranging from liquid crystal displays to data storage and information processing [2].

We report model simulations for a laser-operated chiral molecular switch, namely 1-(2-cis-fluoroethenyl)-2-fluorobenzene. This styrene derivative supports one achiral and two chiral atropisomers, the former being the most stable isomer at room temperature. In quantum wavepacket propagations the isomerization of 1-(2-cis-fluoroethenyl)-2-fluorobenzene is investigated based on electronic ground and excited state potentials obtained from (TD)DFT calculations. Using IR laser pulses the molecule is excited to an internal rotation around its chiral axis, i.e. around the C-C single bond between phenyl ring and ethenyl group, causing a change of the chirality. On the basis of stereoselective laser pulses [3], a laser control mechanism was developed which allows for the selective switching between the achiral and either the left- or right-handed form of the oriented molecule [4], see Fig. 1. To orient the molecule it is bonded to adamantane which on his part serves as rigid molecular tripod to be chemisorbed, e.g. on a gold surface via thiol linker groups [5]. The symmetry of the adsorbent defines the orientation of the molecule which influences the selectivity and efficiency of the control.

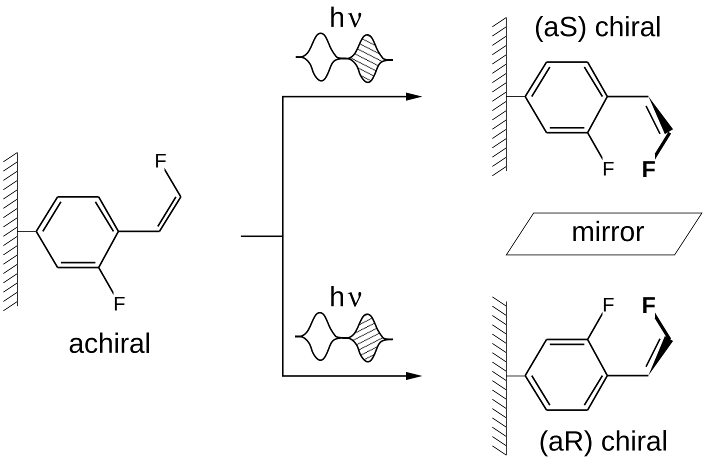

Figure 1: Stereoselective laser pulses control the transformation between the achiral and either left- or right-handed atropisomer of 1-(2-cis-fluoroethenyl)-2-fluorobenzene.

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Taming of a beast: Multi-mode nonadiabatic excited state dynamics in NO_3

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NO_3 is a D_{3h} symmetrical radical of considerable interest in atmospheric chemistry. Furthermore, it poses a number of tremendous challenges for its theoretical treatment, making it a very interesting benchmark system for method development.

The problems of obtaining accurate *ab initio* electronic structure data was solved only recently by large-scale MRCI calculations.[1] The ground and low-lying excited states can now be computed reliably and in excellent agreement with available experiments.[2] This methodology has been used to compute coupled full-dimensional potential energy surfaces (PESs) for the first excited state ($^2E''$). The *ab initio* data is represented by a diabatic potential matrix, accounting for the strong Jahn-Teller (JT) effect in the doubly degenerate excited state and also for mode-mode couplings between different vibrational coordinates. It turns out that higher-order JT couplings are important for a reasonable description of the complicated PESs. The necessary matrix elements were derived recently[3] and lead to excellent fitting results for the strongly anharmonic PESs.

The development of the surfaces enabled us to study the multi-mode nonadiabatic dynamics of the $^2E''$ state that follows photodetachment from NO_3^-. In a first study, only terms up to second order were retained in the potential matrix. Such a vibronic coupling approach is often sufficient to obtain very good spectra. However, the computed detachment spectrum, though in reasonable agreement with experiment, shows significant differences in the details.[4] In the present study higher-order JT and additional mode-mode couplings are included in the dynamics calculations. This leads to a significantly improved agreement with experiment, demonstrating the importance of mode-mode and higher-order couplings.

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Adsorption energy calculations with the method of increments: Application to the adsorption of CO on the CeO_2 110 surface

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Adsorption processes like CO on the CeO_2 110 surface contain two types of interactions: dispersion and classical electrostatics. Dispersion is a part of the electron correlation that is often referred to a long-range and non-local electron correlation. It is by definition not included in the Hartree-Fock (HF) formalism and is usually not incorporated in density functionals due to the local character of the functional as well. (DFT results are published in [1].) The classical electrostatic interaction between CO consists of the interactions of dipole, quadrupole and all higher moments of CO with the surface of ceria. To describe both effect on an equal footing, it is necessary to use highly accurate wavefunction-based correlation methods like the coupled cluster approach.

But a full CCSD(T) treatment of all atoms which are involved in the binding is not feasible with nowadays computers. Therefore the method of increments is used for the problem. Originally it was invented to calculate the correlation energy of extended systems in terms of contributions of localized orbital groups (for a review see [2]). Here for the first time the method of increments is applied to the adsorption of molecules on surfaces. The main idea is an expansion of the correlation energy of the joint system (surface and adsorbate) in terms of correlation contributions of localized orbital groups numbered $i, j, k, ...$

$$E_{\text{corr}} = \sum_i \epsilon_i + \frac{1}{2} \sum_{\substack{ij \\ i \neq j}} \Delta\epsilon_{ij} + \frac{1}{6} \sum_{\substack{ijk \\ i \neq j \neq k}} \Delta\epsilon_{ijk} + \cdots . \tag{1}$$

One group will be the localized orbitals from the adsorbed molecule, the other groups will be the individual atoms in the solid. We have to take as many into account as their correlation contribution is changed due to the adsorbtion process. We calculate the adsorption energy at the CCSD(T) level with the method of increments and compare to MP2 results, which can be performed both within the incremental expansion and within a full correlation calculation.

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Bonding Properties in Elemental Zn and Cd

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The structures of intermetallic phases are not only governed by the number of valence electrons but also by their degree of localization and the transfer of electrons between the constituents. As a consequence, structures can considerably deviate from close packing, a fact that is not yet understood, even for metallic elements like Zn and Cd, which stick out from the hexagonal close packed elements due to the exceptionally large c/a ratio.

The studies presented here combine theoretical and experimental investigations to determine the electronic structure of Zn and Cd in position and in momentum space in comparison to that of Mg, which has formally the same valence electron configuration, but in contrast to Zn and Cd, a nearly ideal c/a ratio.

DFT calculations were performed with various local and gradient corrected functionals including hybrid functionals [1]. Although the anisotropy of the bonding is not described in a well-balanced manner at this level of theory, the role of electronic correlation, involving also the filled d-shell, and the different nature of the intra- and inter-layer interactions can be obviously deduced from the results. The potential energy surface with respect to the lattice constants, PES(a,c), proved to be very flat along a path that corresponds to a uniaxial compression along the c axis. Depending on the method and the computational parameters, even a second local minimum is apparent. Although the band structure and the topology of the Fermi surface varies along this path, leading to a Mg-like appearance at the second minimum, these changes are not reflected in the computed difference electron densities and Compton profiles.

In order to get hints whether the special shape of the potential energy surfaces of Zn and Cd is an artefact of DFT or a real property of the elements, wave-funtion-based methods are applied, computing the correlation energy by means of the method of increments.[2,3] The viability of this approach for group II metals has been demonstrated.[4,5] Preliminary calculation for Zn indeed show a second minimum on PES(a,c), being however separated from the first one by a rather high barrier.

Both, the static deformation densities obtained from multipole refinement of the X-ray diffraction data, and the anisotropy of the measured Compton profiles look similar to the computed DFT-results. However the measured differences between Mg and Zn are more pronounced.

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Titles of posters

[The abstracts can be obtained upon request to the organizers (stc07@mx.uni-saarland.de).]

1. Explicitly Correlated Local MP2-Theory
   T.B. Adler,[1] G. Knizia,[1] O. Marchetti[1], F.R. Manby[2] and H.-J. Werner[1]
   [1]*Institut für Theoretische Chemie, Universität Stuttgart, Stuttgart, Germany*
   [2]*School of Chemistry, University of Bristol, Bristol, United Kingdom*

2. Theoretical and Spectroscopic Study on 2-Substituted 1,3-Indandiones as Potential Optical Sensors for Metal Ions
   Anife Ahmedova[1] and Michael Springborg[2]
   [1]*Faculty of Chemistry, University of Sofia, 1 James Bourchier Blvd., Sofia 1164, Bulgaria*
   [2]*Physical and Theoretical Chemistry, University of Saarland, Saarbruecken 66123, Germany*

3. Simulation study of the structure and dynamics of the Alzheimer's Amyloid beta (Abeta) peptide monomer in aqueous solution
   Priya Anand,[1,2] Ulrich. H.E. Hansmann,[1] and F.S.Nandel[2]
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Germany
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4. The total number of bound states in a given attractive potential
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5. Noncovalent Interactions between Graphene Sheets and in Multishell (Hyper)Fullerenes
J. Antony,[1] C. Mück-Lichtenfeld,[1] and S. Grimme[1]
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6. Assessment of local hybrid functionals for thermochemistry and barriers
H. Bahmann,[1] A.V. Arbuznikov,[1] and M. Kaupp[1]
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7. Correlation factor model
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8. Hidden Dynamical Quasi-Symmetry in Optical Absorption and Ionization Spectra of Quantum Dot Nanorings
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9. Decomposition of Methylamine ($\mathrm{CH_3NH_2}$): Spectroscopy and Kinetics
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10. Vanadium Oxide Compounds with Quantum Monte Carlo
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11. Unbiased study of stochiometric $(CdS)_n$-Cluster
Michael Bauer,[1] Yi Dong,[1] and Michael Springborg,[1]
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12. Metadynamics Simulations of Phase Transitions in Silicon
Using a Novel High-Dimensional Neural Network Potential
J. Behler,[1,2] R. Martoňák,[3] and M. Parrinello[2]
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13. Probing the Photoswitch:
non-adiabatic Hybrid QM/MM MD Studies of Liquid Azobenzene
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14. Theoretical Study of Electrochemical and IR Spectroscopic Properties of Transition Metal Coordinated Scorpionates
J. Borowka,[1] H. Vitze,[1] M. Wagner,[1] and M. C. Holthausen[1]
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15. Modelling tunnel effects in multidimensional quasi-classical trajectories
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16. A modified DMBE IV Potential with spectroscopic accuracy - vibrational levels and rotational constants
J. Brandão[1] and C.M.A. Rio[1]
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17. Molecular dynamics simulations of dendrimer-encapsulated α-Keggin ions in trichloromethane solution
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18. On the Chelate Effect in Uranyl Complexes
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19. Point defects in CeO_2 and CaF_2 investigated using the periodic fast multipole electrostatic embedding scheme
A. Burow,[1] M. Sierka,[1] and J. Sauer[1]
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20. Molecular Dynamics Simulations for a Light-Driven Molecular Motor
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21. MO-based prediction of rate constants for the indirect photolysis of organic compounds
A. Böhnhardt, R. Kühne, R.U. Ebert, and G. Schüürmann
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22. All-electron Douglas-Kroll-Hess and pseudopotential study on the low-lying states of Uranium hydride UH
X. Cao,[1] A. Moritz,[1] and M. Dolg[1]
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23. (s)-N-Acetylproline Amide in Liquid Water: Assessing the Quality of the Continuum Solvation Approach
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24. Metal-Ligand Mo-X (X = P, N, PO, and NO) Bond Situation in Mo Complexes
G.F. Caramori and G. Frenking
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25. Structural and electronic analysis of lanthanide complexes: f orbitals may be not just spectators and reactivity may not necessarily be independent of the identity of the lanthanide atom; a DFT study
H. Chermette,[1] S. Schinzel,[1] and M. Visseaux[2]
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26. Assessment of the Exchange-Correlation Functionals for the Physical Description of Spin Transition Phenomena by DFT Methods: All the Same?
H. Chermette,[1] P. Fleurat-Lessard,[2] S. Zein,[2] S.A. Borshch,[2] and M.E. Casida[3]
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27. Through-bond versus through-space contributions on excitation energy transfer in condensed phase

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28. ROKS simulation of the excited-state dynamics of pyrrole
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29. Molecular and Electronic Architecture of Bimetallic Anionic $[\textit{cyclo}\text{-Cu}_3\text{Au}_3]^-$ Homotops
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30. Hybrid Density Functional Theory Study of Photoluminescence Spectra of Platinum II Porphine and Platinum II Octaethylporphine
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31. Substituted Azobenzene Molecules as Molecular Switches: Quantum Chemical Investigations
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32. Properties of Al_2O_3, AlO, and Al clusters
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33. Decoherence Mechanisms in Nanotechnology
G. Doyen,[1] D. Drakova[2]
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34. The Role of Decoherence for STM induced CO desorption on Cu(111)
D. Drakova,[1] G. Doyen[2]
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35. Efficient Algebraic Evaluation of Second Quantized Operator Strings: Term Generation and Simplification

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36. A Theoretical Investigation on the Influence of the Microhydration on the Vertical and Adiabatic Spectrum of 1-Methylthymine
M. Etinski[1] and T. Fleig[1]
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37. Multi-mode vibronic interactions in fluorinated benzene radical cations
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38. Direct Estimate of Conjugation, Hyperconjugation, and Aromaticity with the EDA Method.
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39. Ultrafast Dissociation Pathways of Diphenylmethyl Chloride to Generate Reactive Carbo Cations
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40. Superexchange and double exchange in diluted magnetic semiconductors
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41. Ab-initio DFT/TDDFT study of manganese porphyrin systems
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42. Implementation of the Incremental Scheme and Application to CCSD Energies
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43. Implementation of the Incremental Scheme for CCSD Energies
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44. Solving ROKS equations in excited state molecular dynamics simulations
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45. First-Principles Calculations of Magnetic Circular Dichroism Spectra
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46. Optimizing the DFTB repulsive energy using genetic algorithms
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47. ^{51}V NMR Chemical Shifts Calculated from QM/MM Models of Vanadium Chloroperoxidase
    K.R. Geethalakshmi, M. Bühl, Mark P. Waller, Dongqi Wang, and Walter Thiel
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48. Understanding Enzymatic Reaction Mechanisms using QM/MM Simulations:
    Hydrolysis of Nucleotides in Membrane-Bound hGBP1
    R. Glaves, A. Chaumont, G. Mathias, and D. Marx
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49. Presence or absence of metal-metal bonding in boron- and carbon-bridged dinuclear transition metal complexes from ELF and AIM studies
    K. Goetz, M. Kaupp
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50. Coupling of Short-Range Density Functional Theory With Long-Range Wavefunction-based Methods: New Results
    E. Goll,[1] H.-J. Werner,[1] and H. Stoll[1]
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51. Binding of Uranyl(VI) and Pertechnetate Ions in Water
    V. Golubnychiy, M. Bühl
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52. Non-adiabatic dynamics on open shell systems: Application to OHF.
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53. Surface states on Xe covered Cu(111)
    V.G. Grigoryan,[1] M. Springborg,[1] F. Forster,[2] and F. Reinert[2]
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54. Theoretical analysis of simple models of nitrogenases based on molybdenum (III). Modification of the electronic environment to modulate the catalytic activity
   P. Guadarrama[1] and H. Vázquez-Lima[1]
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55. Dynamic Protonation Equilibrium of Solvated Amino Acid Side Chain Analogues
   W. Gu,[1] T. Frigato,[1] T.P. Straatsma,[2] and V. Helms[1]
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   [2] *Computational Sciences and Mathematics Division, Pacific Northwest National Laboratory, USA*

56. Quantum dynamics calculations of the ground state tunneling splitting
   in malonaldehyde
   Thorsten Hammer[1] and Uwe Manthe[1]
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57. Application of MRexpT:
   N_2 and LiF as higher excitations of H_4, P_4, BeH_2
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   Greinstrasse 4, 50939 Cologne, Germany

58. Investigation of some truncated Coupled Cluster Variants
   A. Hansen, F. Wennmohs, and F. Neese
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59. Reactions of Transition Metal Carbenes With Ethylene
   R. Haunschild and G. Frenking
   Fachbereich Chemie, Philipps-Universitt Marburg, 35032 Marburg, Germany

60. Quantized density-functional theory for molecular fluids in nanoporous materials
   Serguei Patchkovskii,[1] Sergei N. Yurchenko,[2] and Thomas Heine[2]
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61. Restriction of Vibrational Coupling Contributions in Molecular Cluster Calculations
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62. Benchmarking the performance of spin-component scaled CC2
   in ground and electronically excited states
   A. Hellweg,[1] S.A. Grün,[1] and C. Hättig[1]
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63. Efficient implementation of recent developments in MP2-F12 theory and its application to chemical problems
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64. Interaction of probe molecules with the metal organic framework (MOF) CPO-27-Ni examined by periodic DFT calculations
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65. Quantum chemical calculations on Ni_2O_2 and $Ni_2O_2^+$
O. Hübner[1] and H.-J. Himmel[1]
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66. Vibronic Structure Methods for Larger Molecular Systems:
Theoretical Prediction on Photoelectron Spectra of RNA-Building Blocks
J. Huh and R. Berger
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67. Exploring the Photo-induced Green-to-Red conversion of Fluorescent Protein EosFP
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68. Molecular parity violation in open-shell systems
within a electroweak quasirelativistic approach
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69. Molecular Dynamics investigations on DJ-1 protein and its mutants associated with Parkinson's disease
A. Jezierska,[1,4] F.E. Herrera,[1] S. Gustincich,[2,3] and P. Carloni[1]
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70. Dynamical and Static Models of n-Butylboronic Acid - Comparison with Experiment
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71. First principle molecular dynamics simulations of selected model structures of hydrogen-bonded Schiff and Mannich bases - comparison with experimental data
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72. Atomistic Modelling of Mechanical Properties of Silicon
Hanno Kamp[1] and Michael Springborg[1]
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73. Unraveling the Effect of NH_3 co-Ligation on the Catalytic Efficiency of Fe^+ Ion in Gas Phase Carbon Dioxide Fixation Reactions
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74. Deposition of Ni_{13} and Cu_{13} clusters on Ni(111) and Cu(111) surfaces
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75. Transition strength and first-order properties of excited states from local CC2 for large molecules
Danylo Kats,[1] Tatiana Korona,[2] and Martin Schütz[1]
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76. Tracing Photochemical Products of NBA by Resonance Raman Spectroscopy
K. Kiewisch,[1] S. Laimgruber,[2] P. Gilch,[2] and J. Neugebauer[1]
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77. Parallelization of large-scale relativistic multi-reference configuration interaction programs with application to $(RbBa)^+$ and the thorium diatom
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78. Static electric dipole polarizabilities for closed shell atoms and ions
    V. Koch, D. Andrae
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79. Calculation of excitation energies employing the TPSS meta-GGA
    S. Kossmann and F. Neese
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80. Tuning the reactivity of $ZnO(10\bar{1}0)$ surfaces by coadsorption of CO_2 and characterization of the adsorbate structures by STM data calculations
    R. Kovacik, B. Meyer, and D. Marx
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81. On the elliptic relation between physical and mathematical entanglement
    T. Krüger[1]
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    Institut für Chemie, Karl-Franzens-Universität Graz, 8010 Graz, Austria

82. Quantum-Chemical Study on the Effect of Lewis Acid Centers in a Poly(ethylene oxide)-Based Solid Electrolyte
    P. Kubisiak and A. Eilmes
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83. Second order Møller-Plesset treatment of molecular hydrogen physisorption on MOF-5
    A. Kuc,[1] G. Seifert,[1] T. Heine,[1] and H. Duarte,[2]
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84. Unbound exciton-phonon states in oligothiophene crystals – a model approach for spectroscopic purposes
    W. Kulig[1] and P. Petelenz[1]
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85. Application of *short-range*-DFT/ *long-range*-Coupled-Cluster method to Lanthanide Diatomics
    S. Chabbal[1] and T. Leininger[1]
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86. Open-Shell Local Coupled Cluster Theory
    Y. Liu and H.-J. Werner
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87. Raman Optical Activity Spectra: Analysis of the Role of the Electric-Dipole–Electric-Quadrupole Tensor
    S. Luber ,[1] C. Herrmann,[1] and M. Reiher[1]

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88. 2,2'-Bipyridyl Ytterbocene Complexes: Molecular Kondo Singlet Analogs?
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89. The Density Matrix Renormalization Group Method Facing the Spin State Problem in Quantum Chemistry
K. H. Marti,[1] G. Moritz,[1] I. Malkin Ondik,[1] and M. Reiher[1]
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90. Analytic High-Order Douglas–Kroll–Hess Electric Field Gradients
R. Mastalerz,[1] G. Barone,[2] R. Lindh,[3] and M. Reiher [1]
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91. Ab initio study of hydrogen storage in MOFs
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92. DFT/Metadynamics Study of the Methanol Synthesis on ZnO Catalysts
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93. Simple Gaussian Model Density to Describe Electrostatic QM/Pot Interaction
F. Mohamed[1] and J. Sauer[1]
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94. Relativistic Energy-consistent 5f-in-core Pseudopotentials for Penta- and Hexavalent Actinides
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95. Towards the chemistry of element 112 (eka-mercury). Benchmark relativistic calculations of properties of E112 vs. Hg compounds.
N.S. Mosyagin,[1] A.V. Titov,[1] A.N. Petrov,[1] T.A. Isaev,[1] A.V. Zaitsevskii,[2] and E.A. Rykova[3]
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96. Parity Violating Nuclear Magnetic Resonance Parameters: Scaling Laws and Conformational Effects
    S. Nahrwold,[1] M. Bühl,[2] and R. Berger[1]
    [1] *Frankfurt Institute for Advanced Studies, Johann Wolfgang Goethe University, Frankfurt am Main, Germany*
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97. Docking with PSO@Autodock: A Novel Fast Docking Program Employing the *var*CPSO-ls Particle Swarm Algorithm
    Vigneshwaran Namasivayam, Robert Günther
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98. Methodological Developments in the Theory of the Zero-Field Splitting Tensor
    Frank Neese
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99. Photochemistry of nucleobases: An nonadiabatic *ab initio* molecular dynamics study of uracil in aqueous solution
    H. Nieber and N. L. Doltsinis
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100. Molecular Magnetism: An *ab initio* study of a hexanuclear ferric wheel
     H. Nieber,[1,3] K. Doll,[2,3] and G. Zwicknagl[3]
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101. Hydrogen halides photochemistry on small water particles: Joint theoretical and experimental study
     M. Ončák,[1,2] P. Slavíček,[1,2] M. Fárník,[2] V. Poterya,[2] and U. Buck[3]
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102. Electrophilic Reactivity of Michael-Type Acceptors
     Norbert Ost,[1] Alexander Böhme,[1] Ralf-Uwe Ebert,[1] Ralph Kühne,[1] Gerrit Schüürmann[1,2]
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103. Computational structural investigations of bilirubin binding motif analogues
     J.J. Panek,[1] M. Novič,[1] S. Passamonti[2]
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104. An unexpected requirement for the DFT treatment of agostic interactions: the uniform electron gas correlation limit.
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105. Theoretical studies of excitation energy transfer between two benzene molecules
J. Pfister,[1] R.F. Fink,[1] and B. Engels[1]
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106. Pigment Yellow 101: An example for the rich photochemistry of medium-sized molecules
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107. Joint theoretical and experimental study of the water/corundum (Al_2O_3) interface: An application to the safety assessment of nuclear waste disposal
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108. ^{31}P Chemical Shift Tensors in RNA and DNA Backbone. A DFT Study of Conformational Dependence and Its Implications for NMR Studies
J. Přecechtělová, P. Padrta, M.L. Munzarová and V. Sklenář
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109. Lagrangian approach to molecular vibrational Raman intensities
D. Rappoport[1] and F. Furche[1]
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110. Structures and energetics of pure and mixed acetylene-ammonia clusters
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111. Vibrational properties of adsorbates on surfaces
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112. A theoretical modeling of cysteamine solutions Raman spectra
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113. A String-Based Implementation of Explicitly-Correlated Coupled-Cluster Theory
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114. Embedded Cluster calculation on the ZnO-Catalyzed Synthesis of Methanol from CO and H_2
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115. HYLLERAAS-TYPE WAVE FUNCTIONS FOR ATOMS AND SMALL MOLECULES
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116. QM/MM Study of BlrB: a BLUF Domain Protein
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117. QM/MM Studies on Adsorption Compelxes in Zeolites
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118. A new method for correlated dynamics of many electron systems with reduced density matrices
B. Schäfer-Bung, M. Nest, and P. Saalfrank
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119. An improved algorithm for the calculation of non-expanded second-order exchange contributions in the framework of the combined density functional and symmetry-adapted perturbation theory approach
R. Schäffer, G. Jansen
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120. Phenol$(H_2O)_{1\leq n\leq 3}$ revisited: An *ab-initio* study on the photophysics at the level of Coupled Cluster response theory

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121. Representation of the eigenfunctions of $\hat{L}^2$ with one dimensional basis functions

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122. Joint theoretical and experimental research on actinide compounds

     B. Schimmelpfennig,[1] R. Klenze,[1] and K. Gompper[1]

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123. Density-Functional Study of Electronic Structure and EPR Parameters of Biologically Relevant Manganese Complexes

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124. Rotational excitation and cross sections in the reaction $Cl^- + CH_3Br \rightarrow ClCH_3 + Br^-$

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125. An anisotropic Heisenberg model on the trigonal lattice for multiferroic oxides

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126. Improvement of trial wave functions for quantum Monte Carlo

     A. Schwarz,[1] R. Berner,[1] and A. Lüchow[1]

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127. Gold Nanoparticles Without and With Ligands

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128. Prediction of Hydrogen Bond Donor and Acceptor Strengths of Organic Compounds

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129. Structures and energies of mono-sulphated dimers from carrageenans by DFT method
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130. Electronic structure of excited electronic states of tryptophan and N-acetyl tryptophan methyl amide (NATMA)
D. Shemesh and W. Domcke
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131. Are six isomers involved in folding if tryptophan
is one of the amino acids in the peptide?
D. Shemesh and E.W. Schlag
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132. Theoretical investigation of the heavy-atom analogues of acetylene E2X2 (E = Si-Pb, X=F-I)
Takayasu Shimizu and Gernot Frenking
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133. A Combined DFT and High Resolution 27Al NMR Study of Isolated Aluminum Atoms in the Framework of ZSM-5 Zeolite
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134. Quantumchemical studies of the adsorption of single acetone molecules on hexagonal (Ih) and cubic (Ic) ice polymorphs.
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135. How much can donor/acceptor-substitution change the responses of long push-pull systems to DC fields?
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136. The extended Algebraic Diagrammatic Construction scheme of second order (ADC(2)-x) for Open-Shell Molecules: A Study of Polyene Cations and Polyenyl Radicals
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137. Many-Electron Bands in Transition Metal Compounds
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138. Implementation of a vector potential method in an *ab initio* Hartree-Fock program
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139. Monte Carlo Simulation of Temperature Programmed Desorption Including Binding Energies and Frequency Factors Derived by DFT Calculations
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140. A Refined Unsymmetric Lanczos Eigenvalues for Computing Accurate Eigentriplers of a Real Unsymmetric Matrix
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141. Theoretical Studies of homogeneous SiGe clusters and their comparison with pure Si and Ge clusters using Density Functional Tight binding (DFTB) method
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142. Moment-constrained two-step Density Fitting in periodic local MP2 (DFP-LMP2). Application to molecular crystals and adsorption on surfaces.
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143. Infrared Spectroscopy and Dynamics of the Protonated Water-Dimer
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144. Theoretical study of the C-H activation mechanisms with uranium(IV) and thorium(IV) bismethyl complexes

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145. A Comparative Study of Single Reference Correlation Methods of the Coupled-Pair Type

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146. Modeling Biological $\mathrm{U}^{\mathbf{VI}}$ Coordination from First Principles

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147. Theoretical study of adsorption of methanol and toluene on zeolite surfaces

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148. Equilibrium structures of the adenine thymine base pair
in the ground and first excited state

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149. Comparing Ring Polymer and Centroid molecular dynamics methods

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150. Transition States of the H Abstraction from Amines through Reaction with OH Radicals

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151. A Local Version of the Algebraic Diagrammatic Construction Scheme to Calculate Electronic Excited States of Large Molecules

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152. The dynamics of angular degrees of freedom: new basis set and grid representations of Hamiltonian operators

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153. Combination of Spectroscopy and Quantum Chemistry in Elucidating the Reaction Mechanisms of Nonheme Iron Enzymes
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154. Hydrogen storage in nanoporous materials
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155. Beyond the featureless particle approximation: An extension of quantum-liquid density-functional theory to account for rotational effects
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156. Theoretical Investigation of the Molecular Properties of Cerium Trichloride $CeCl_3$
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157. Theoretical estimation of Zero-Field Splitting Tensor
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158. STM-induced Switching of Hydrogen on a Silicon(100) Surface:
An Open-System Density Matrix Study
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159. *Ab initio* Calculation of Excitation Energy Transfer between two Perylene Bisimide Molecules
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160. Antitumor activity of a copper(II) chelate complex. An approach in the explanation of the mechanism of action through molecular modelling and docking studies

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161. Absorption Spectra Simulation for Small Donor-Acceptor Molecules with Density Functional Method
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162. Assessment of the High Accuracy Extrapolated Ab Initio Thermochemistry (HEAT) method for the computation of ionization potentials.
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163. α,$\delta-$Hybrid Peptides as Peptide Mimetics
Peter Schramm,[1] Carsten Baldauf,[2] Gangavaram V.M. Sharma,[3] and Hans-Jörg Hofmann[1]
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164. Analytical Gradients in a Subsystem Formulation of Density Functional Theory
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3 News from UK's CCP9 Programme

UK's Collaborative Computational Project 9 (CCP9) on ”Computational Studies of the Electronic Structure of Solids”

3.1 Report on the CCP9 Conference

3.1.1 Report on Psi-k/CCP9 Workshop on ”Solving the Bogoliubov-de Gennes and Gross-Pitaevskii equations for superconductors, superfluids and BEC”

Manchester

September 29-30

Sponsors: Psi-k and CCP9

W.M. Temmerman and James Annett

Organisers Report

The Psi-k/CCP9 workshop on ”Solving the Bogoliubov-de Gennes and Gross-Pitaevskii equations for superconductors, superfluids and BEC” was held at the Britannia Hotel Manchester over the weekend of 28-30 September 2007. This is was the eighth workshop on this theme, following previous workshops held in Bristol, most recently in 2003.

The workshop was organized around three focussed themes: ab initio theory of superconductivity, Bose condensation and BEC in ultracold atomic gasses, and nanoscale superconductivity. The ab initio theory of superconductivity session was led by a review of the current state of the art in DFT for superconductors, by E.K.U. Gross. His introduction was followed by discussion of the probable spin triplet superconductor, strontium ruthenate, by K.I. Wysokinski. The microscopic origin of d-wave pairing in the 2d Hubbard model, as found in Quantum Monte Carlo Dynamical Cluster Approximation was presented by T., Maier. Surprisingly it was shown that while d-wave pairing occurs in the one band Hubbard model, multiband models show pairing in some cases but not others. Finally F. Mauri described calculations of electron-phonon coupling and superconductivity in intercalated graphites from first-principles.

The session on atomic BEC and other Bose condensates was opened by M. Szymanska who presented evidence for room temperature BEC in a polariton system. She described how the polaritons have a short lifetime and so require continuous pumping, therefore the theory leads to a new type of condensation which can exist in a highly non-equilibrium and dissipative environment. F. Marchetti described the BEC to BCS crossover effects in spin polarized Fermi gases, and analogues to magnetic superconductors. R. Scott presented computational dynamical models of optical effects such as reflection, diffraction, focusing and interferometry produced with Bose-Einstein condensates. He showed that this atom optics is directly analogous to traditional optics, but there are important differences due to atom-atom interactions, scattering events and finite temperature phase fluctuations. A new approach to stochastic phase space methods to study the effect of quantum and thermal fluctuations on bosonic atom dynamics in optical lattices was presented by J. Ruostekoski. The stochastic simulations were able to produce the observed damping rate of the atomic center-of-mass motion within the experimental measurement uncertainty of recent NIST experiments. The session on nanoscale superconductivity began with a talk by I.V. Grigorieva. She presented recent experimental findings, where vortices in mesoscopic superconductors were observed directly by the Bitter decoration method. It was found that the circular geometry of superconducting disks leads to the formation of concentric shells of vortices and the filling of the shells is governed by well defined rules ("periodic law"). with 'magic' vortex numbers corresponding to the appearance of consecutive new shells. F. V. Kusmartsev described mechanisms for nanoscale structure formations in underdoped High Temperature superconductors taking into account both phonons and strong correlations. Doping leads to layered growth of clusters and such specific charge density waves around random, charged impurities continues with decreasing density, and the formation of the multi-shell droplets divides the charged boson fluid into two phases: bosons localized in clusters and superfluid bosons. He also reported on exciting recent experiments indicating production of THz radiation from layered High T_c superconducting devices. A. Kormanyos discussed Andreev edge channels and magnetic focusing in normal-superconductor hybrid systems: for which he presented a semiclassical conductance formula including both effect of electron-like as well as hole- like quasiparticles. in the Andreev reflection process. J.P. Hague used analytical results and quantum Monte-Carlo simulations to show how a combination of lattice geometry, long-range attraction and local repulsion can lead to pairs which are both small and light. These small-light pairs are a precursor to high condensation temperatures, providing a model for high T_c superconductivity. Finally J. Robinson reported a systematic study of Nb/FM/Nb trilayer structures in which the FM layer is one of the strong ferromagnets: Co, Fe, Ni, or $Ni_{80}Fe_{20}$(Py). Inconsistencies were found between the exchange energies derived elsewhere from S/FM bilayer experiments and from S/FM/S junction measurements compared to their bulk Curie temperatures. A Josephson effect 0 to Pi phase-transition was found in Co based superconducting junctions which shows evidence for the appearance of a second-harmonic in the current-phase relation at the minimum of the critical current. In summary the workshop showed deep and interesting connections between the phenomena of superconductivity in metals, both conventional and exotic systems like high Tc cuprates, and corresponding phenomena in atomic condensates and physically very different systems such as BEC of polaritons. The mechanisms of pairing via strong correlation, and BEC optical lattice analogues of the Mott Hubbard physics of strongly correlated materials remain an exciting area of developments. The experimental ability to control devices and materials on the nanoscale also leads to links with many problems relating to spintronics and magnetism in multilayer systems which are central to Psi-k and CCP9 activities.

Programme

Friday 28 September:

Arrival

Saturday 29 September:

Ab initio theory of superconductivity 9.00-9.45
E.K.U. Gross (Berlin)

Superconducting phases in strontium ruthenate subject to magnetic field 9.45-10.30
K.I. Wysokinski (Lublin)

Coffee 10.30-11.00

Dynamic cluster simulations of the 2D Hubbard model: What is the cuprate pairing mechanism? 11.00-11.45
T. Maier (ORNL)

Electron-phonon coupling and superconductivity in intercalated graphites from first-principles 11.45-12.30
F. Mauri (Paris)

Lunch 12.30-2.00

BCS-BEC in strongly dissipative systems 2.00-2.45
M. Szymanska (Oxford)

Polarised Fermi Superfluids: Phase Diagram and Dynamics 2.45-3.30
F. Marchetti (Oxford)

Tea 3.30-4.00

Atom optics with Bose-Einstein condensates 4.00-4.45
R. Scott (Nottingham)

”BECs in time-periodic external fields” 4.45-5.30
T.S. Monteiro (UCL)

Banquet - Yang Sing 7pm

Sunday 30 September:

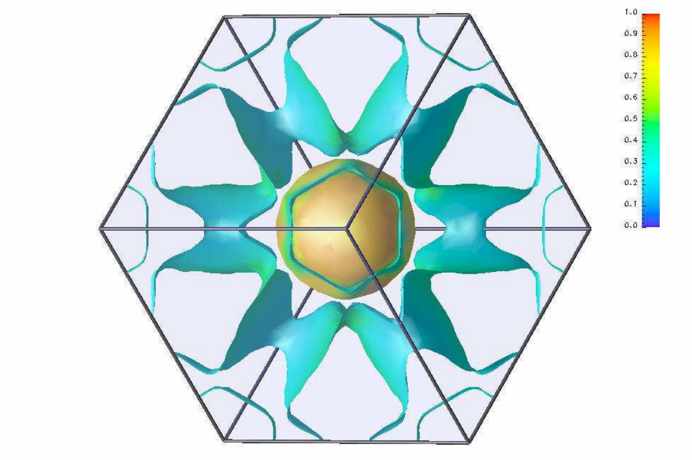
Non-equilibrium dynamics beyond Bogoliubov - de Gennes: Dissipative bosonic atom dynamics in optical lattices using stochastic phase space methods J. Ruostekoski (Southampton)	9.00-9.45
Vortex configurations in nanostructured superconductors: direct observations using Bitter decoration I. Grigorieva (Manchester)	9.45-10.30
Coffee	10.30-11.00
Role of impurities in Superconductor-Insulator Transition F. Kusmartsev (Loughborough)	11.00-11.45
Andreev edge channels and magnetic focusing in normal-superconductor hybrid systems: a semiclassical analysis A. Kormanyos (Lancaster)	11.45-12.00
Superlight small bipolarons in the presence of a strong Coulomb repulsion Jim Hague (Loughborough)	12:00-12:15
A systematic study of strongly ferromagnetic S/FM/S Pi-junctions Jason Robinson	12:15-13:00
Lunch	13:00

Abstracts

”Ab-initio theory of superconductivity”

E. K. U. Gross (Berlin, Germany)

A novel density-functional-type approach to the description of phonon-mediated super-conductivity is presented [1]. The goal of this approach is to provide a theory with predictive power, allowing the calculation of material-specific properties such as the critical temperature.To this end, the electron-phonon interaction and the electron-electron repulsion are treated on the same footing. There are no adjustable parameters such as the μ^* of Eliashberg theory. The formalism can be viewed as the superconducting generalization of the multi-component density-functional theory [2] for electrons and nuclei. Approximations of the universal xc functionals are derived on the basis of many-body perturbation theory [1,3]. Numerical results for the critical temperature and the gap will be presented for simple metals [4,5], for MgB_2 [6], and for Li, Al and K under pressure [7]. In particular, for MgB_2, the two gaps and the specific heat as function of temperature are in very good agreement with experimental data. For Li and Al under pressure, the calculations explain why these two metals behave very differently, leading to a strong enhancement of superconductivity for Li and to a clear suppression for Al with increasing pressure. For K we predict a behavior similar to Li, i.e. a strong increase of T_c with increasing pressure. Furthermore, the peculiar features of the superconducting phase of CaC_6 will be analyzed [8]. In the figure below, the gap function of CaC_6 is shown on the Fermi surface. Finally, results for hydrogen under extreme pressure will be presented. It turns out that hydrogen is a three-gap superconductor whose critical temperature increases with increasing pressure until about 100K (at 500 GPa).



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"Superconducting phases in strontium ruthenate subject to magnetic field"

Karol Izydor Wysokinski (Lublin, Poland)

The orbital symmetry of the spin triplet superconducting state of strontium ruthenate is widely believed to be chiral one with d-vector along the crystal c-axis. This piece of evidence comes mainly from measurements of the spin susceptibility being constant below T_c when measured in magnetic field in a-b plane. The experimental measurements of the spin susceptibility in a magnetic field along c-axis show that small magnetic field is sufficient to rotate d-vector into the a-b plane. Using the realistic 3 dimensional and 3 band model of the superconductor in tight -binding representation we solve the Bogolubov-deGennes equations and discuss the effect of the (spin-only) magnetic field on the material. The model predicts field induced phase transition characterized by small but finite entropy change thus explaining the lack of its experimental signatures.

”Dynamic cluster simulations of the 2D Hubbard model: What is the cuprate pairing mechanism?”

Thomas A. Maier (ORNL, USA)

We will discuss recent dynamic cluster approximation studies of the doped two-dimensional Hubbard model of the high-temperature superconductors. The phenomena found in these simulations include antiferromagnetic, superconducting and pseudogap behavior, and are thus remarkably similar to what is observed in the cuprate materials. We will discuss how one can combine diagrammatic techniques with numerical results to obtain the momentum, frequency, temperature and spin quantum number that characterize the pairing interaction responsible for d-wave pairing. The effective pairing interaction is found to be attractive between singlets formed between nearest neighbor sites and its dynamics is associated with the antiferromagnetic spin fluctuation spectrum. An exact decomposition of the pairing interaction reveals that it is mediated by antiferromagnetic spin fluctuations. Motivated by these results, we will then discuss a simple spin susceptibility representation of the pairing interaction that provides potential routes to experimental verification. Furthermore, we will present preliminary results for the frequency dependence of the superconducting gap parameter and discuss recent results for a three-band Hubbard model obtained from a combination of density functional and dynamic cluster quantum Monte Carlo calculations.

”Electron-phonon coupling and superconductivity in intercalated graphites from first-principles”

F. Mauri

Graphite intercalated compounds (GICs) can display a superconducting behavior at low temperature. However, until the discovery of Yb and Ca intercalated graphite [1,2] (Tc(YbC_6)=6.5 K and Tc(CaC_6)=11.5 K), the critical temperatures achieved were typically less than a Kelvin. Recently, it has been shown that even higher critical temperatures (up to 15.1 K) can be achieved in CaC6 applying hydrostatic pressure [3]. This is presently the highest Tc reported in a GIC. The large number of intercalant available for graphite (more than 100) requires theoretical investigation to guide the experimental work. Moreover since there is not a general consensus on the nature of the superconducting state, the calculation of the superconducting properties in the framework of strong-coupling electron-phonon theory for different intercalants and the consequent comparison with experiments can judge its reliability in predicting the critical temperature. Using density functional theory we investigate the occurrence of superconductivity in AC_6 with A=Mg,Ca,Sr,Ba. All the aforementioned GICs posses an intercalant band at the Fermi level, GICs[4]. Comparing the superconducting properties and the calculated critical temperatures of these materials we show that the pairing mechanism is due to the elenctron-phonon interaction. In particular we find CaC_6 to be superconducting with T_c=11.5, and we predict Ba and Sr intercalated graphite to be superconducting with T_c 0.2 K and 3.0 K [5]. Our predictions have been confirmed by recent experiments [6]. We study the pressure dependence of T_c and and that the SrC_6 and BaC_6 critical temperatures should be substantially enhanced by pressure. On the contrary, for CaC_6 we find that in the 0 to 5 GPa region, Tc weakly increases with pressure. The increase is much smaller than what shown in several recent experiments. We propose possible explanations for these discrepancies. Finally we argue that, although MgC_6 is unstable, the synthesis of intercalated systems of the kind $Mg_xCa_{(1-x)}C_6$ could lead to higher critical temperatures.

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”BCS-BEC in strongly dissipative systems”

Marzena Szymanska (Oxford, UK)

By confining photons in a semiconductor microcavity, and strongly coupling them to electronic excitations, one may create polaritons: bosonic quasi-particles with an effective mass of 10^-9 times that of Rubidium atoms, thus allowing BEC at elevated temperatures. After a long and strenuous search, the comprehensive set of experiments has finally given evidence for BEC of polaritons [1]. However, this new condensate depart from the archetypal BEC in several ways. Most importantly polaritons have short lifetime and so a continues pumping is required to sustain a steady-state. This leads to a new type of condensation which can exist in highly non-equilibrium and dissipative environment [2,3].

We show that, surprisingly, the mechanism of condensation, connected with the chemical potential reaching the bottom of bosonic modes, is the same in closed systems at equilibrium and in open systems with pump and decay. In the latter, the role of the chemical potential is played by the energy at which the non-thermal distribution diverges. We derive the self-consistency condition for an uniform condensed steady state. This condition can be compared both to the laser rate equation and to the Gross-Pitaevskii equation of an equilibrium condensate. However, even when the system is characterised by a thermal distribution, the presence of pumping and decay significantly modifies the spectra of phase fluctuations: the low energy phase mode (Goldstone, Bogoliubov mode) becomes diffusive at small momenta, leading to correlation functions – and thus condensate lineshape – that differ both from an isolated condensate and from that for phase diffusion of a single laser mode. These generic features of dissipative condensation can be observed in different optical probes of the microcavity and affect the decay of spatial and temporal coherence in a condensate, and modify its superfluid properties.

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"Polarised Fermi Superfluids: Phase Diagram and Dynamics"

Francesca Marchetti (Oxford, UK)

Recent advances in the ability to manipulate and control ultra-cold atomic vapours have provided a unique experimental system in which to explore pairing phenomena. Following the successful realisation of the crossover from the BCS state of Cooper pairs to the Bose Einstein condensation of diatomic molecules, attention has turned to the consideration of more exotic Fermi superfluids. A subject that has attracted particular theoretical and experimental interest is that of Fermi condensates with imbalanced spin populations, owing to the potential relevance of polarised Fermi condensates to QCD and magnetised superconductors. I will discuss the topology of the zero and finite temperature phase diagram for both cases of equal and unequal masses [1,2], the relevance of our findings to recent experiments, and the dynamics of phase separation [3].

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"Atom optics with Bose-Einstein condensates"

Robin Scott (Nottingham, UK)

Optical effects such as reflection, diffraction, focusing and interferometry, which are traditionally created with laser light, can also be produced with Bose-Einstein condensates. In many ways atom optics is directly analogous to traditional optics, but there are important differences due to atom-atom interactions, scattering events and finite temperature phase fluctuations. We show that these additional effects can have a pronounced effect on the dynamics of the atom cloud, sometimes spoiling the atom optics, but occasionally enhancing it, for example in interferometry. We also explain the different techniques we use to capture the key underlying physics.

BECs in time-periodic external fields

T.S. Monteiro, UCL

I will review experimental and theoretical work on the dynamics of cold atoms and BECs subjected to time periodic driving. At the non-interacting limit, coherent matter wave dynamics of atoms subjected to short pulses from sinusoidal optical lattices represents a clean experimental test of quantum chaotic dynamics[1]; new possibilities arise for manipulating cold atoms [2] leading to an experimental demonstration of the first fully coherent quantum ratchet [3]. In the presence of interactions, recent experiments mean the dynamical stability of the condensate is of much importance. We investigate the mechanism for destabilization using propagation of the GPE and the time-dependent Bogoliubov approach[4]. In certain quantum many- body Hamiltonians, AC-driving offers the possibilities for manipulating the superfluid-Mott Insulator transition[5] and quantum state transfer in spin-chains[6].

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Non-equilibrium dynamics beyond Bogoliubov - de Gennes: Dissipative bosonic atom dynamics in optical lattices using stochastic phase space methods

J. Roustekoski (Soton, UK)

We study the effect of quantum and thermal fluctuations on bosonic atom dynamics in optical lattices using stochastic phase space methods. As a first example, we consider the dipolar motion of bosonic atoms in a very shallow, strongly confined 1D optical lattice [1] using the parameters of the recent NIST experiment [2]. We find that, due to momentum uncertainty, a small, but non-negligible, atom population occupies the unstable velocity region of the corresponding classical dynamics, resulting in the observed dissipative atom transport. The stochastic simulations are able to produce the observed damping rate of the atomic center-of-mass motion within the experimental measurement uncertainty.

As a second example we study the splitting of a harmonically trapped atomic Bose-Einstein condensate when we continuously turn up an optical lattice [3,4]. As the lattice height is increased, quantum fluctuations of atoms are enhanced. We analyze the resulting nonequilibrium dynamics of the fragmentation process of the condensate, the loss of the phase coherence of atoms along the lattice, and the reduced atom number fluctuations in individual lattice sites. The comparisons to the atom number squeezing experiments [5] are made. We find the atom number squeezing to saturate in deep lattices due to nonadiabaticity in turning up of the lattice potential that is challenging to avoid in experiments when the occupation number of the lattice sites is large, making it difficult to produce strongly number squeezed or the Mott insulator states with large filling factors.

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”Vortex configurations in nanostructured superconductors: direct observations using Bitter decoration”

I.V. Grigorieva (Manchester, UK)

Vortices in superconductors represent a system of interacting particles that can be easily tuned and controlled by the applied field or a specially designed pinning potential. Confining the vortices in a mesoscopic (i.e., no more than a few microns in size) superconductor provides yet another tool to force them into specific configurations, control their number or even make vortices to merge into multiquanta flux lines with a single normal core (giant vortices). I will present our recent experimental findings, where vortices in mesoscopic superconductors were observed directly by the Bitter decoration method. In particular, we found that the circular geometry of superconducting disks leads to the formation of concentric shells of vortices and the filling of the shells is governed by well defined rules ("periodic law"). We were also able to identify 'magic' vortex numbers corresponding to the appearance of consecutive new shells. Furthermore, we investigated the effect of pinning on vortex configurations in confined geometry: While weak pinning only leads to distortions of the vortex shells, strong pinning forces up to 10 vortices to merge into tight clusters or, for sufficiently strong pinning, even true giant vortices. The experimental findings are in good agreement with the results of numerical simulations based on Ginzburg-Landau equations.

Role of impurities in Superconductor-Insulator Transition

F. V. Kusmartsev[1] and M. Saarela[2]
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We investigate the Nanoscale Structure Formations in underdoped High Temperature superconductors taking into account both phonons and strong correlations. The phonons may be responsible for local pairing that may lead to a formation of bi- tri- and other string and stripy polaron structures. The quantum nature of these structures and the strong Coulomb interaction force us to take into account many body quantum correlations.

The formation of superconducting and insulating states in underdoped cuprates may be well described by a two-dimensional strongly-interacting charged boson gas. This is one of the most fundamental and clean quantum systems. At high densities and zero temperature it is superconducting whereas at very low densities it forms a Wigner crystal where all charged carriers are localized and the system is an insulator. The existence of oxygen impurities with opposite charge equal to -2e lead to a formation of charged structures or the charge density waves symmetrically distributed around the impurities. Moreover, with decreasing density the Coulomb attraction of the impurity collects a cluster of bosons around it and induces oscillations of the boson density in the tail part. When the density decreases the amplitude of these oscillations increases. At the critical density, r_s=3, a small cluster of four particles forms a bound state with the impurity. Outside this localized cluster or "an artificial atom" formed by correlations a fraction of positive background is revealed to keep to whole system neutral.That "atom" will attract the next shell of boson when the density is further decreased.

The layered growth of clusters and such specific charge density waves around random, charged impurities continues with decreasing density. The formation of the multi-shell droplets divides the charged boson fluid into two phases: bosons localized in clusters and superfluid bosons. Obviously, when the radius of those clusters is significantly smaller than the distance between impurities the flow of bosons through the system is superfluid. However, when the distance between impurities becomes equal to the cluster diameter then bosons will be fully localized. Such a state is no more superconducting even though some superconducting islands still exist. This is a quantum phase transition which has the first order character. Such a transition reminds the percolation phase transition with the difference that the main role here is played by the over-screening of the Coulomb interaction.

We present detailed results on this system using variation many body method based on the Jastrow type correlated wave function and show that they provide the microscopic explanation for the recent experimental findings in superconducting-insulator transition in high T_c superconductors [1,2] and formation of various structures observed in STM experiments[3].

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”Andreev edge channels and magnetic focusing in normal-superconductor hybrid systems: a semiclassical analysis”

Andor Kormányos (Lancaster, UK)

We study a transverse electron-hole focusing effect in a normal-superconductor system. The spectrum of the quasiparticles is calculated both quantum mechanically and in semiclassical approximation, showing an excellent agreement. A semiclassical conductance formula is derived which takes into account the effect of electron-like as well as hole-like quasiparticles. At weak magnetic fields the semiclassical conductance shows characteristic oscillations due to the Andreev reflection, while for stronger fields it goes to zero. These findigs are in line with the results of previous quantum calculations and with the expectations based on the classical dynamics of the quasiparticles[1].

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”Superlight small bipolarons in the presence of a strong Coulomb repulsion”

J.P. Hague (Loughborough)

Through the use of analytical results and quantum Monte-Carlo simulations, we show how a combination of lattice geometry, long-range attraction and local repulsion can lead to pairs which are both small and light. In particular, such pairs can be found for reasonable magnitudes of the phonon frequency and electron-phonon coupling. The possible Bose-Einstein condensation of the pairs is discussed. We show that small-light pairs are a precursor to high condensation temperatures. The stability of such a condensate to phase separation and disorder are also discussed.

”Systematic study of strongly ferromagnetic S/FM/S π-junctions”

J. Robinson (Cambridge, UK)

We report a systematic study of Nb/FM/Nb trilayer structures in which the FM layer is one of the strong ferromagnets: Co, Fe, Ni, or $Ni_{80}Fe_{20}$(Py) [1,2]. Accurate control of the FM layer thickness has enabled detailed studies of the magnetic and transport properties in the superconducting state. In all cases we estimate the thickness of the magnetic dead layer and the exchange energies of the ferromagnetic layers; in doing so we demonstrate inconsistencies between the exchange energies derived elsewhere from S/FM bilayer experiments and from S/FM/S junction measurements compared to their bulk Curie temperatures which may hint at further complexity in the underlying physics. Finally, we focus in detail on a single 0 to π phase-transition in Co based superconducting junctions and show evidence for the appearance of a second-harmonic in the current-phase relation at the minimum of the critical current [2,3].

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4 General Workshop/Conference Announcements

4.1 11th International Symposium on Physics of Materials (ISPMA) 11

Prague, Czech Republic

24-28 August, 2008

The next International Symposium on Physics of Materials (ISPMA) 11 will take place in Prague, Czech Republic, on 24 - 28 August 2008, two days after the NANO SPD 4 Conference in Goslar, Germany.

Since 1972, the ISPMA Series has brought together colleagues working all over the world in the field of Physics of Materials and Materials Science and Technology.

The next Symposium ISPMA 11, organised in cooperation with the Metals Science Society of Czech Republic, will focus on fundamental and applied aspects of physics of metals and alloys, metal hydrides, intermetallics, non-metallic materials, composites, submicrocrystalline and nanocrystalline materials, and materials science.

A special session will be devoted to materials prepared through severe plastic deformation.

Please visit

http://material.karlov.mff.cuni.cz/events/ispma11

to see the first announcement on the ISPMA 11.

For further information on the ISPMA series, the website of the ISPMA 10 (2005) at

http://material.karlov.mff.cuni.cz/events/ispma10/

has been still available.

You can check also the ISPMA 10 Proceedings appearing in

Materials Science & Engineering A, Volume 462, Issues 1-2, Pages 1-476 (25 July 2007), International Symposium on Physics of Materials, 2005, edited by F. Chmelik, R. Kral, J. L. Martin and G. Kostorz

http://www.sciencedirect.com/science/journal/09215093 .

Please forward this messages to your colleagues who might also be interested in visiting the ISPMA 11.

Frantisek Chmelik
Premysl Malek
(Conference Chairs)

Regina Cerna
(Conference Secretary)

4.2 DPG08: Symposium on ”Ferroic materials and novel functionalities”

German Physical Society Meeting

25 - 29 February, 2008

Berlin, Germany

We would like to draw your attention to the upcomming symposium on

”Ferroic materials and novel functionalities”

at the annual meeting of the German Physical Society meeting in Berlin, on 25 - 29.02.2008

http://nanoscience.physik.uni-halle.de/dpg/

The focus will be on complex oxide materials, especially new phenomena at their surfaces and interfaces. Parallel to the symposium there will be a contributed session with an identical title in Section Surfaces (Oberflächen) and you are most wellcome to submit contributions for that.

The Organizers,

Ingrid Mertig, Rossitza Pentcheva, Silvia Picozzi and Wolfgang Kleemann

4.3 International Workshop: Quantum Monte Carlo in the Apuan Alps IV

Sat 26th July - Sat 2nd August 2008
The Towler Institute
Vallico Sotto, Tuscany, Italy

www.vallico.net/tti/tti.html (Institute)
www.vallico.net/tti/qmcitaa_08/announcement.html (Event)
www.vallico.net/tti/qmcitaa_08/poster.png (Event Poster)

Continuing the series of alternative and very informal meetings at this venue, the Cambridge University Theory of Condensed Matter group is organizing a fourth International Workshop to discuss the development and application of the continuum quantum Monte Carlo method in condensed matter physics and quantum chemistry. The conference will take place in a 16th Century monastery in the mediaeval high mountain village of Vallico Sotto (in the Tuscan Apuan Alps near the beautiful Italian city of Lucca).

The normal format for these events involves formal presentations being restricted to the mornings, with the afternoons left free for relaxed discussion and participation in fun activities. For the young and vigorous, we organize mountain walks, caving and other healthy outdoor exercise, whilst the unfit and elderly might enjoy artistic tours, city visits and spa treatments, with all participants reuniting in the evening for excellent Tuscan dinners in local restaurants. The monastery is a unique venue where the community spirit and magnificent location have inspired memorable meetings in the past.

This year's workshop will involve up to 35 people, all accommodated on site. Given the limited space most speakers will be specifically invited, but anyone who feels that they have something to contribute and who wishes to attend the event is most welcome to contact the organizers (Mike Towler: mdt26 at cam.ac.uk) for further details. There is no charge either for attendance at the conference or accommodation.

Confirmed speakers: David Ceperley, Neil Drummond, Matthew Foulkes, Pablo Lopez Rios, Lubos Mitas, Richard Needs, Norbert Nemec, John Trail, Mike Towler.

FURTHER DETAILS AND PHOTOS FROM PREVIOUS WORKSHOPS ACCESSIBLE ON TTI WEB PAGE - CLICK THE 'PUBLIC EVENTS' LINK OR LOOK AT 'EVENT PHOTOS'. TTI CURRENTLY TAKING BOOKINGS FOR THE HOSTING OF CONFERENCES, SCHOOLS AND GROUP MEETINGS IN EASTER/SUMMER 2008. ENQUIRIES WELCOME.

4.4 Summer School: Quantum Monte Carlo and the CASINO program III

Sun 3rd August - Sun 10th August 2008
The Towler Institute
Vallico Sotto, Tuscany, Italy

www.vallico.net/tti/tti.html (Institute)
www.vallico.net/tti/qmcatcp_08/announcement.html (Event)
www.vallico.net/tti/qmcatcp_08/poster.jpg (Event Poster)

The third international summer school in the series ‘Quantum Monte Carlo and the CASINO program’ will take place during August 2008 at the Towler Institute monastery in the Tuscan Apuan Alps, organized and hosted by members of Cambridge University physics department’s Theory of Condensed Matter (TCM) Group. The aim of the school is to give students a thorough introduction to quantum Monte Carlo as a method for performing high-quality calculations of the electronic structure of real materials. The course is designed for young scientists who have no previous experience with this technique, though anyone interested is welcome to take part.

Describing the complex behaviour of materials at the atomic level requires a sophisticated description of the correlated motion of the electrons. Quantum Monte Carlo (QMC) is an increasingly popular and explicitly many-body method with the unusual capability of yielding highly accurate results whilst also exhibiting a very favourable scaling of computational cost with system size. Over the last seventeen years, the Cambridge group have been researching QMC methods and we have created a powerful, general computer program - CASINO - to carry out the calculations. The school will focus both on the basic theory of QMC and on more advanced practical techniques, and will include a thorough introduction to the CASINO program. A background in density functional theory or similar is normally thought to be useful.

The monastery is a delightful 16th century building incorporating an ancient church, and is situated in the spectacular isolated Tuscan mountain village of Vallico Sotto. It is fully equipped with state-of-the-art presentation and computer technology. All accommodation is on-site. There is a strictly-enforced policy for all events at the Institute that considerable time should be set aside for mountain hikes, caving, canyon walking, swimming, eating in great local restaurants, visits to classical concerts in Lucca and Barga, and similar activities.

Instructors will include the main authors of the CASINO program, (Richard Needs, Mike Towler, Neil Drummond, and Pablo Lopez Rios) and possibly others. The cost of the school for one week including accommodation will be around 400 GBP. Participants would need to arrange a flight to Pisa airport from where onward transportation will be arranged. Interested students should contact Mike Towler (mdt26 at cam.ac.uk) for registration and further details.

FURTHER DETAILS AND PHOTOS FROM PREVIOUS SCHOOLS ACCESSIBLE ON TTI WEB PAGE - CLICK THE ‘PUBLIC EVENTS’ LINK OR LOOK AT ‘EVENT PHOTOS’. TTI CURRENTLY TAKING BOOKINGS FOR THE HOSTING OF CONFERENCES, SCHOOLS AND GROUP MEETINGS IN EASTER/SUMMER 2008. ENQUIRIES WELCOME.

4.5 Conference on Molecular Simulations in Biosystems and Material Science

April 2 - 5, 2008

University of Konstanz, Germany

We have the pleasure to announce the Conference on Molecular Simulations in Biosystems and Material Science (April 2 - 5, 2008) of the ESF-programme SIMBIOMA at the University of Konstanz (Germany).

Similar to the SIMU-conference in 2001 (Konstanz), the conference in 2008 will be organized in 9 plenary talks (50 minutes), 10 sessions with three speakers (invited talks with 30 minutes talks and contributing talks with 20 minutes), short poster presentation talks (3 minutes each), and in addition a general poster session.

The topics of the sessions include:

* Proteins and Peptides
* Protein Crystallization and Aggregation
* Membranes
* Quantum Effects in Nano-sized Materials
* Microfluidics
* Soft Matter I
* Soft Matter II
* Methods
* Multiscale Methods
* Charge Transfer and Transport in Condensed and Soft Matter

Time and spatial scales relevant for material science and biosystems are challenges in the fundamental modeling of matter. In this conference we would like to bring together computational scientists facing these challenges and to focus the subject on new techniques as well as on current applications for bridging the gap between ab-initio and meso and macroscopic levels. The conference will aim at developing a real cooperation among the groups participating in the SIMBIOMA ESF program. It is also open to participants, who are not members of this program.

We invite you to have a look at the web page of the Conference for details on the list of invited speakers, preliminary programme, registration procedure and abstract submission:

http://www.uni-konstanz.de/simbioma/

For further information, please contact: simbioma@uni-konstanz.de

Please note the following deadlines:

* December 15, 2007: Poster abstracts submitted up to this date will be reviewed by the Advisory Committee for the selection of contributed talks (20 min). Abstracts submitted later cannot be considered as contributed talks.

* February 20, 2008: End of abstract submission for poster contributions and of regular registration

FOR SIMBIOMA MEMBERS ONLY:

On behalf of the organizing committee we invite you (as a qualified member of the ESF-SIMBIOMA- programme) or a co-worker of your laboratory indicated by you to participate and present at least a poster in this general conference of our SIMBIOMA ESF programme. In order to continue the SIMBIOMA- programme successfully it is necessary that every `‘‘SIMBIOMA-participant_laboratory’’` sends at least one member to this conference. To help your participation we can provide the following support.

For you (or the participating co-worker from your laboratory) the registration fee is waived and the lunch meals and the banquet will be paid by us. Concerning the local costs SIMBIOMA can perhaps offer you a support (up to about EUR 120), depending on available funds and requests.

We would enjoy very much if you could accept our invitation but would like to insist that you send at least one person from your lab.

Prof. Peter Nielaba

Conference Advisory Committee:

A. Alavi, M. P. Allen, W. Andreoni, M. Athenes, J.-L. Barrat, K. Binder, P. Bolhuis, M.E. Cates, G. Ciccotti, C. Dellago, M. Dijkstra, I. Economou, P. Espanol, R. Everaers, P. Faisca, D. Frenkel, D. Galli, H. Grubmueller, B. Hafskjold, J.P. Hansen, K. Jacobsen, H. Jonsson, K. Kremer, S. Lago-Aranda, E. Leontidis, B. Lesyng, R. Lipowsky, D. Mackernan, D. Marx, V. Mohacek-Grosev, O. G. Mouritsen, R. Netz, P. Nielaba, L. Nilsson, M. Rodger, U. Roethlisberger, J. P. Ryckaert, M. Scheffler, F. Schmid, B. Smit, M. Sprik, S. Succi, J. Ulicny, I. Vattulainen, M. Vendruscolo, R. Vuilleumier, M. Waroquier

Organizing Committee:

W. Andreoni, G. Ciccotti, P. Nielaba, B. Smit, R. Vuilleumier

Local Organizing Committee:

P. Nielaba, Y. Fischer (Conference Secretary), Members of P. Nielaba’s Group

5 General Job Announcements

PhD/Postdoctoral Position for ”Multiscale Modeling of Mechanical Properties of Interfaces”

Institute of General Materials Properties, Department Materials Science and Engineering
University Erlangen-Nuernberg, Germany

The Institute of General Materials Properties, in the Department Materials Science and Engineering of the University Erlangen-Nuernberg, Germany, has a vacant a PhD / postdoc position in the group of Prof. Alexander Hartmaier, which is to be filled as soon as possible.

The focus of the research work lies on the mechanisms of grain boundary sliding in metals. Grain boundary sliding plays an important role in the deformation and fracture of nanostructured and ultrafine-grained materials, especially at high temperatures. To contribute to the advancement of the design and development of such novel nanomaterials, state-of-the-art computational methods will be applied. Particularly, accurate ab initio calculations will promote the qualitative and quantitative understanding of the stability of grain boundaries based on the atomic bonding across the interface. The gained knowledge will be used within continuum methods to study the mechanical behavior of larger structures.

Prof. Alexander Hartmaier's simulation group consists of young and motivated members with a strong expertise in computational methods and numerical modeling. The methods as well as the hardware equipment used are state-of-the-art. In addition to the numerical investigations, the group is also active in experimental studies, relating microstructure to the mechanical properties of nanomaterials.

The Department Materials Science and Engineering has a long tradition in materials science. It is situated in Erlangen, a lively baroque town in a beautiful region of Frankonia. In addition to the activities of the University of Erlangen-Nuernberg, research and development in the area is brought forward by institutions such as the Fraunhofer Institute for Integrated Systems, Siemens, and other innovative companies.

The ideal candidate should have a profound background in solid state physics or materials science, extensive experience with either electronic structure calculations or continuum models, and a strong interest in microstructure and mechanical properties of materials.

The position is limited to two years in the first phase, but can be prolonged for two further years. There is the possibility to obtain a doctoral degree. For further information or submission of your complete application please contact

Dr. rer. nat. Rebecca Janisch
University Erlangen-Nuernberg
Department Materials Science
Institute I: General Materials Properties Martensstr. 5
91058 Erlangen
Germany

E-mail: rebecca.janisch@ww.uni-erlangen.de
Phone: +49 (0) 9131 85 27473
Fax: +49 (0) 9131 85 27504

The application must contain a CV, a list of publications, a statement of research interests, and two letters of recommendation. Application deadline is November 30th 2007.

The University of Erlangen-Nuernberg is an affirmative equal opportunity employer.

Two postdoctoral and one Ph.D. position in southern France on ”New Tools for the Simulation of Solids and Interfaces”

The hosting labs are Centre de Physique Moléculaire Optique et Hertzienne (Bordeaux) and Institut pluridisciplinaire de recherche sur l'environnement et les matériaux (Pau).

This reseach project is motivated (i) by synthesis work and simulation studies on dyes grafted on surfaces at the university of Pau, (ii) by a novel algorithm to calculate molecular spectra developed at the university of Bordeaux and (iii) by expertise on real time computer steering also available at Bordeaux.

The overall aim is to couple the molecular dynamics code dl_poly with the DFT code Siesta in order to provide a QM/MM package for solids and interfaces; to apply a new TDDFT algorithm for the calculation of molecular spectra; to provide tools for visualization and steering of running, coupled, parallel dl_poly+Siesta jobs and to provide new molecular mechanics force-fields for simulation of adsorption in zeolites or on surfaces.

Candidates for the three positions below will be expected to interact closely with colleagues providing experimental data for the validation of the calculations.

The following positions are available:

1: Co-supervised Ph.D. position on: **QM/MM treatment of localized excited states in organic-inorganic hybrid materials and application to photo sensitizers grafted on silica.**

Ph.D. advisors : R. Brown (Pau) and D. Foerster (Bordeaux).

Starting : December 2007 , duration : 3 years, net Salary : 1200 euros. The position is based in Pau, with travel to Bordeaux and short internships with developers of the component codes in Daresbury, UK and San Sebastian in Spain. Candidates should have a Master's degree or equivalent in scientific computing and quantum and classical molecular modelling.

2: Postdoctoral position on:

Calculation of optical spectra of large molecules from TDDFT linear response, using a new algorithm

Collaborators : D. Foerster (Bordeaux) and I. Baraille(Pau).

Starting: January 2008, duration: 1 year, (renewable twice), net salary ca. 2100 euros. The position is based in Bordeaux, with travel to Pau and contact with developers of the siesta code at San Sebastian in Spain. Candidates should be familiar with scientific computing and density functional theory.

3: Post-doc position on:

Parametrization of an empirical force field for the silica surface from two dimensional periodic ab initio DFT calculations

Collaborators : A. Marbeuf (Bordeaux) and I. Baraille(Pau).

Starting : December 2007, duration : 1 year (renewable once), net salary ca. 2100 euros. Candidates should have experience in the ab initio modelling of periodic systems. Experience in molecular dynamics simulation would be appreciated.

Interested candidates are encouraged to contact I. Baraille, isabelle.baraille@univ-pau.fr and provide vitae, list of publications and letters of recommendation.

POSTDOCTORAL POSITIONS AT MATERIALS SIMULATION LABORATORY

Physics Department, University of South Florida, USA

The Materials Simulation Laboratory (MSL) at USF Physics Department invites applications for several postdoctoral positions which are available immediately. The MSL research interests include simulations of matter at extreme conditions, molecular electronics, spintronics and development of methods for large scale atomistic simulations of materials. For more information please visit MSL web site at

http://msl.cas.usf.edu

Applicants should have a PhD degree in theoretical/computational condensed matter physics, chemistry, materials science and other closely related fields. The successful candidates should also have extensive programming skills in C/C++, Fortran and Linux environment. Knowledge parallel programming including MPI is a plus.

Interested applicants should send a CV, complete list of publications, 1-2 page description of research experience and technical skills, names and email addresses of 3 references to Prof. Ivan Oleynik at iioleynik@gmail.com .

Two Postdoctoral Positions

Computational Materials Group at University of Wisconsin in Madison

http://matmodel.engr.wisc.edu/

Postdoctoral Position in Continuum Level Modeling of Diffusion through Complex Microstructures for Nuclear Applications

We invite applications for a postdoctoral position in the Computational Materials Group at the University of Wisconsin Madison. The objective of this project is to determine relationship between microstructure and diffusion in materials used for TRISO fuel coatings. The research is a multiscale project, building from atomic to microstructure length scales with a combination of ab initio, molecular dynamics, kinetic Monte Carlo, and continuum simulation. There is also close interaction with experimental collaborators at UW-Madison. The candidates work will primarily involve continuum level modeling of transport but they will work closely with a team of PhD students to construct the multiscale model. A PhD in materials science, physics, or related field is required. Familiarity with both atomic scale and continuum modeling is preferred but not required.

The appointment is jointly with Professors Izabela Szlufarska and Dane Morgan, initially for one year with possible extension. Candidates should send a detailed CV (preferably in pdf format) and contact information for 3 references to Izabela Szlufarska at izabela@engr.wisc.edu. Review of applicants will begin immediately and will continue until the position is filled.

Postdoctoral Position in First-Principles Based Modeling of Alloys for Nuclear Applications

A postdoctoral position is available immediately in the Computational Materials Group at the University of Wisconsin in Madison. The research will be in the general area of combining First-Principles (DFT) and Monte Carlo methods to model thermodynamics, diffusion, and evolution of radiation damage in multicomponent alloys of interest for nuclear applications. Other materials applications may also be pursued part time for interested applicants. Familiarity with DFT methods and alloy thermodynamic and kinetic modeling is essential.

The appointment is with Professor Dane Morgan, initially for one year with possible extension. Interested applicants should send CV (preferably in PDF format) and contact information for 3 references to Dane Morgan at ddmorgan@wisc.edu. Review of applicants will begin immediately and will continue until the position is filled.

Computational Materials Group (CMG) (http://matmodel.engr.wisc.edu/): CMG is a joint effort of Izabela Szlufarska and Dane Morgan in the Department of Materials Science and Engineering at UW Madison. We are a highly active group, with 16 members and work in areas including nanomechanics, nano-bio interfaces, energy materials, and geophysics.

Dane Morgan
Assistant Professor
244 Materials Science & Engineering
1509 University Avenue
Madison, WI 53706-1595
Phone: 608-265-5879, Fax (department): 608-262-8353
Email: ddmorgan@wisc.edu
Web: `http://www.engr.wisc.edu/mse/faculty/morgan_dane.html`

Postdoctoral Fellowship
Oak Ridge National Laboratory, USA

A postdoctoral fellowship is available to work with David Singh at Oak Ridge National Laboratory.

Project Description:

Postdoctoral fellow sought to use first principles and first principles based methods to understand materials properties with particular emphasis on transport and dynamics, and thermal and thermoelectric properties such as thermal conductivity and mechanisms for thermal conductivity reduction, thermoelectric power, and electron-phonon coupling.

Qualifications: Ph.D. degree required in physics or other physical sciences. Experience in the use of first principles methods for materials desired.

Applicants cannot have received the most recent degree more than five years prior to the date of application and must complete all degree requirements before starting their appointment.

How to Apply:

Qualified applicants may apply online at

`https://www2.orau.gov/ORNL_POST/`.

All applicants will need to register before they can begin the online application. For complete instructions, on how to apply, please see the instructions at

`http://www.orau.gov/orise/edu/ornl/ornl-pdpm/application.htm`.

When applying for this position, please reference the position title and number (ORNL07-80-MSTD).

POSTDOCTORAL RESEARCH ASSOCIATE

HOWARD UNIVERSITY-BOEING CORPORATION RESEARCH COLLABORATION

Department of Electrical and Computer Engineering, Howard University

Washington, D.C., USA

A postdoctoral position is available immediately to work with Prof. Steven L. Richardson in the Department of Electrical and Computer Engineering at Howard University in Washington, D.C. on a joint Howard University-Boeing Corporation Research Collaboration on the Electronic Structure of Polymeric Materials. Successful candidates should have experience in quantum chemistry or electronic structure theory and possess either a Ph.D. in chemistry, physics, or materials science. Strong communications skills for writing papers and making presentations are required and a considerable degree of independence as a professional researcher is expected of the candidate.

Funding for this project is for at least one year with the possibility of renewal for a second year. The postdoctoral research associate will be physically located at Howard University in Washington, D.C. Because this is a university-industry collaboration applicants should either be U.S. citizens or possess a green card.

Applicants should provide: (i) an updated curriculum vitae, (ii) complete list of publications, and (iii) the name and contact information for three references. All of this information should be sent electronically to Prof. Steven L. Richardson (srichards22@comcast.net) as soon as possible.

Postdoctoral and Ph. D. Positions in Donostia (ETSF) Spain

The Nano bio Spectroscopy Group and the local node of the ETSF invites applications for two postdoctoral positions and one PhD position. (http://dipc.ehu.es/arubio)

The research projects will directly address fundamental problems related to the characterization of the electronic properties of complex systems. Topics to be addressed are:

* Theoretical description of charge transport in molecular systems

* Quantum dots and nanotubes for quantum information applications.

* Nanostructured materials for photovoltaic devices

* Postdoc position on applications to complex systems with biological interest:

Our purpose is to understand the specific process of light emission by the chromophore. This will allow us to extract information about its 'in-vivo' conformation and how its environment, inside the protein, leads to changes in the emission properties (color and intensity). The latter problem suggests the investigation of new ideas beyond present TDDFT/QM/MM implementations. Our last objective is to pave the way for a completely new 'divide and conquer' strategy, with the aim of incorporating the dynamical coupling between the optically active part of the protein, the protein itself, and the surroundings (solvent, substratum, etc.)

The work will be conducted in close relation with the theoretical groups of the NANOQUANTA network of Excellence (www.nanoquanta.eu) as well as within the global framework of the European Theoretical Spectroscopy Facility (ETSF) (http://etsf.eu)

Applicants must have, or expect to obtain before the start of the work, a Ph.D. in a relevant discipline. A strong background in theoretical condensed matter science, chemistry or biology and experience with electronic structure methods are essential, and a knowledge of bio-materials or biophysics is desirable. Good knowledge and experience with DFT as well as in the development and implementation of numerical approaches are highly appreciated.

We consider applications until the positions are filled. Please send your application (either by e-mail or postal mail) as soon as possible to

Dr. Angel Rubio
European Theoretical Spectroscopy Facility (ETSF)
Dpto. Fsica de Materiales UPV/EHU
Edificio Jose Mari Korta
Avd Tolosa 71
20018 San Sebastian, SPAIN
Phone : +34-943018292
Fax : +34-943015600
E-mail: Angel.Rubio@ehu.es

http://dipc.ehu.es/arubio

NOTE: Type of appointment: contract as visiting research professor (for the postdoctoral position). The appointment os for two years with possibility of extension.

Postdoctoral Position in Aarhus, Denmark

Postdoctoral position in development and application of Hybrid Density Functional Theory at University of Aarhus A one-year post-doctoral position (with possibility for prolongation a second year) starting no later than March 1st, 2008 will soon be announced in the group of Bjrk Hammer at the interdisciplinary nano-science center (iNANO), Department of Physics and Astronomy, University of Aarhus in the field of Density Functional Theory. The position is part of a collaboration between physics and computer science at University of Aarhus and Atomistix A/S.

The successful candidate will be responsible for the implementation, testing and application of state of the art hybrid functionals. The successful candidate will work in a dynamic and growing group and will be expected to interact with both computer scientists and scientist applying DFT. We seek a researcher with a proven record in the field of computational modeling preferably with prior experience with DFT simulations. Experience with code development will be an advantage. Applicants must hold a PhD degree in Physics, Chemistry, Chemical Engineering, or Materials Science.

The salary and appointment terms are consistent with the current collective agreement for Danish Universities. The monthly salary is approx. Euro 3300 depending on the level of experience.

If you are interested in the position, please e-mail Georg Madsen (georg.madsen@inano.dk) as soon as possible, including a curriculum vitae, a list of publications, and the names, affiliations, and e-mail addresses of three references. Please e-mail only plain text or pdf-files. Please quote "Position 317k" in the subject header of the email.

Postdoctoral Position in Computational Mineral Physics

Nanochemistry Research Institute, Curtin University of Technology, Perth, Western Australia

A post-doctoral position is available within the Computational Group of the Nanochemistry Research Institute of Curtin University for 2 years. Water is present in nominally anhydrous minerals (NAMs) as hydrogen defects, often at very low concentrations. However, this water has profound influence on the physical, chemical and mechanical properties of these NAMs. The aim of the project is to model the interaction of water with impurities and extended defects in Mg_2SiO_4 forsterite to gain an insight at the atomic level, into the influence of water on deformation mechanisms. The Nanochemistry Research Institute (NRI) is a Tier 1 research institute of Curtin University and consists of a team of both experimental and theoretical researchers working in fields including minerals, crystal growth, sensors, functional materials and energy storage. More information can be found on our web site

http://www.nanochemistry.curtin.edu.au/.

In 2008, the NRI will be relocating to a new purpose built laboratory as part of the major development of a Resources and Chemistry Precinct. The computational group, comprising Profs. Kate Wright, Andrew Rohl, Leslie Glasser, and headed by Prof. Julian Gale, is involved in development of methods and software, such as GULP and SIESTA, as well as in research on a wide range of solid-state applications.

Qualifications: Applicants should have a PhD in mineral physics, solid state chemistry or physics and have experience of ab initio and/or classical solid state modelling methods.

Duration: 2 years

Start date: from January 2008

Location: Nanochemistry Research Institute, Curtin University of Technology, Perth, Western Australia.

Perth is a city of approximate 1.25 million people situated on the Indian Ocean and has a Mediterranean climate. A competitive salary and relocation package is available. Interested candidates should contact Prof. Kate Wright (kate@ivec.org) who will be happy to provide more information.

Postoctoral Position at Rutgers University, USA

A postdoctoral position with Professor Gabriel Kotliar at Rutgers University to develop realistic approaches to the electronic structure strongly correlated materials is available.

We are looking for an individual with a strong backround in computational physics and a strong motivation to develop novel computational tools to predict the physical properties of strongly correlated materials.

We are looking with someone with experience in the development of large scale electronic electronic structure programs. Knowledge of many body physics, GW methods, Dynamical Mean Field Theory , LDA+DMFT, quantum chemistry or molecular dynamics are definitely a plus but are not required.

Applications and three letters of references should be sent by email to Professor Gabriel Kotliar

kotliar@physics.rutgers.edu

Applications will be consider as they arrive and can start either in September 2008 or at an earlier date.

Gabriel Kotliar
Board of Governors Professor of Physics
Department of Physics and Astronomy
Office 271
Serin Physics Laboratory
Rutgers University
136 Frelinghuysen Road
Piscataway
NJ 08854-8019
USA
Phone 732-445-4331
Email: kotliar@physics.rutgers.edu
Fax: 732-445-4343
`http://www.physics.rutgers.edu/~kotliar/`
Secretary: 732-445-4277
Serin Lab Ofice 269

PhD Position

Center for Theoretical Chemistry, Ruhr-University Bochum, Germany

A PhD position in theoretical chemistry is immediately available at the Ruhr-University in Bochum. The work will involve the use of density-functional methods and the development of modern methods for the construction of potential-energy surfaces. The potentials will be applied to describe complex structures of metal clusters at oxide surfaces used in heterogeneous catalysis. The work will be embedded in the Collaborative Research Center SFB558 "Metal - substrate interactions in heterogeneous catalysis" of the German Research Foundation (DFG) (http://www.sfb558.de). A close collaboration with experimentalists at the faculty of chemistry is provided.

The candidate will work in a motivated team and will benefit from the excellent working environment and computational equipment of the Center for Theoretical Chemistry.

Please send your application including CV (preferably by email) to

Dr. Jörg Behler
Lehrstuhl für Theoretische Chemie
Ruhr-Universität Bochum
D-44780 Bochum
Germany
Email: joerg.behler@theochem.rub.de

Consideration of candidates will begin immediately and continue until the position is filled. The University particularly welcomes applications from women.

Max Planck Junior Fellow/Group Leader

Fritz Haber Institute, Max Planck Society, Berlin, Germany

We invite qualified candidates to apply for the position

Head of an Independent Junior Research Group
(Max Planck Junior Fellow)

at the Theory Department of the Fritz Haber Institute (FHI) of the Max Planck Society (MPS) in Berlin, Germany. As the Department has close ties with the the University of California at Santa Barbara, parts of the group's research activities can also be conducted there.

The setting and contract conditions for Independent Junior Research Groups are as follows:

– The group leader position (W2; approximately equivalent to associate professor)
– Postdoc position
– 1 or 2 PhD scholarships
– Technical Assistant position as far as necessary
– Competitive startup package for instrumentation
– Adequate yearly budget for running costs.

An initial 5 year contract will be offered, with possible extension.

The group leader is encouraged to apply for funds from the Deutsche Forschungsgemeinschaft, from ESF and EU, NSF, to host postdocs paid by the Humboldt Foundation, and more.

Independent Junior Research Groups are a high-level award scheme of the Max Planck Society to promote the career of excellent young scientists (emerging leaders). Candidates are expected to show a strong potential to initiate and conduct frontier research in their particular scientific field. Typical candidates are about 3-6 years after their Ph.D. exam.

The ideal candidate will complement the activities of the FHI Theory Department in the field of first-principles theories and statistical mechanics in the wide context of computational materials science and engineering, as well as theoretical biophysics. For an overview of current projects please see our web pages

http://www.fhi-berlin.mpg.de/th .

Applications must be submitted in the official call by the Max Planck Society which will appear in November. Further information and application instructions are given here

http://www.snwg.mpg.de/ .

Deadline for applications is December 14, 2007.

Please feel free to contact Matthias Scheffler to discuss your possible candidature (scheffler@fhi-berlin.mpg.de) or Martin Fuchs (fuchs@fhi-berlin.mpg.de).

Ph. D. Studentship

Thomas Young Centre (TYC), Imperial College London, U. K.

A PhD studentship is available in the TYC at Imperial College London for someone who can start by April 2008 to work on electronic structure calculations of perovskite materials for application as cathodes in solid oxide fuel cells. Anyone interested should please send me or Professor Alan Atkinson (alan.atkinson@imperial.ac.uk) their CV with covering letter.

Professor M. W. Finnis
Department of Materials
and Department of Physics
Imperial College London
Exhibition Road
London SW7 2AZ
U. K.

Email: m.finnis@imperial.ac.uk
Tel: +44 (0) 207 59 46812
Fax: +44 (0) 207 59 46757

Postdoctoral Position in Condensed Matter Physics

”Development of multiscale simulations schemes and applications to material science”

University of Lyon 1, France

A post-doc position is available for one year at the Laboratoire de Physique de la Matière Condensée et Nanostructures, University of Lyon 1 (France). The applicant will work in the modeling group on the development and application of the LOTF technique, a QM/MM molecular dynamics scheme. This technique has been applied to brittle fracture and dislocation dynamics in silicon. Further developments will be carried on by the successful candidate to :

(1) extend the application of the method to polyatomic systems and
(2) use the LOTF technique as an acceleration scheme for the geometry optimization in ab-initio simulations.

Possible applications will concern fracture mechanics, mechanical properties of amorphous systems and the study of platinum-carbon interfaces.

The applicants should hold a Ph. D. in physics and possess a good background in solid state physics. Coding skills in F90 and/or C++ programming languages are required as well as the will to be involved in scientific code development.

This work is founded by the French government through the ANR framework and it includes collaborations with others theoretical groups in Grenoble and Paris.

The net salary after the application of all taxes range between 1900-2100 euros per month.

Applicants should send to the contact below a detailed CV and one or two representative publications.

Contact :

Tristan Albaret
Laboratoire de Physique de la Matière Condensée et Nanostructures
UMR5586 Université Lyon 1/CNRS

Email : tristan.albaret@lpmcn.univ-lyon1.fr
Phone : +33(0)4 72 43 15 65
Fax : +33(0)4 72 43 26 48

Group website :

`http://lpmcn.univ-lyon1.fr/theorie/site/index.php`

ANR (LN3M) website :

`http://www-drfmc.cea.fr/sp2m/L\_Sim/LN3M/index.html`

Postdoctoral Position
University of California, Berkeley, USA

One postdoctoral position may be available in the group of Prof. Ivo Souza in the Department of Physics at the University of California, Berkeley, to begin as early as December 15th 2007. The target area is the *ab-initio* study of phenomena arising from the interplay between broken time-reversal symmetry and spin-orbit interaction. Ongoing work includes studies of the anomalous Hall effect and magnetic circular dichroism in metals. For a list of recent publications on these topics, please see

`http://smilodon.berkeley.edu/publ.html`

Experience with first-principles density-functional methods is essential. Interested applicants should apply by email (`isouza@berkeley.edu`), using "*Post-doctoral search*" as the subject line. Please attach a CV (PDF format preferred) and list names and contact information of 2–3 references. Consideration of candidates will begin immediately and will continue until the position is filled. Potential applicants please consult

`http://smilodon.berkeley.edu/`

for the most up-to-date information on the search.

Ph. D. and Postdoctoral Positions
Katholieke Universiteit Leuven, Belgium

A postdoctoral position is available at the Katholieke Universiteit Leuven (Belgium) to work on transparent conductive oxides (TCO) with regular DFT and GW corrections. Experience with the GW formalism is a strong advantage, as well as the ability to work independently. The position is for one year, and should start in January 2008 or shortly thereafter.

A Ph. D. position is available at the Katholieke Universiteit Leuven (Belgium) to work on transparent conductive oxides (TCO) with regular DFT and GW corrections. Previous experience with ab initio methods is not strictly necessary, but a strong motivation and a good knowledge of condensed matter physics are mandatory. The position is for 4 years, and should start in January 2008 or shortly thereafter.

More information on the scientific topic, the work content and related issues can be obtained from Stefaan.Cottenier@fys.kuleuven.be.

Web site of the K.U.Leuven: http://www.kuleuven.be/english

POSTDOCTORAL RESEARCH ASSISTANT IN THEORETICAL NANOMAGNETISM

Departments of Mathematics: City University, London + Open University,

Milton Keynes, UK

Applications are invited for the post of Postdoctoral Research Assistant in theoretical physics, to work on a project carried out jointly between City University London and the Open University Milton Keynes under the leadership of Professor J. Mathon and Dr A. Umerski. The successful candidate will undertake research into the theory of quantum transport of charge and spin across a ferromagnet-semiconductor interface. This is one of the fundamental problems in the exciting emergent field of spintronics, the area for which the physics Nobel Prize was awarded this year. The proposed research is part of a collaborative EPSRC funded project involving Cambridge University where a parallel experimental investigation will be carried out. Close liaison with the experimental group of Professor Bland in Cambridge is an essential part of the project and frequent visits between the institutions are planned. The successful candidate will also be able to interact with a PDRA and a graduate student working on a closely related EPSRC funded project, Spin@RT, involving some forty researchers based in eight UK institutions (see http://www.spinart.org for details). A PhD in a relevant area of theoretical physics or materials modelling is required and experience in using/modifying/developing electronic structure codes is highly desirable.

The post is a three year fixed term position, available from January 1st 2008, on the RA1A scale with starting salary of £29,448 p.a. inclusive of London Allowance. Informal enquiries should be addressed to either Prof. J. Mathon at j.mathon@city.ac.uk or Dr A. Umerski at A.Umerski@open.ac.uk. Candidates should send a covering letter, together with copy of their curriculum vitae and a list of publications, by 15th January 2008, to

Prof. J. Mathon.
School of Engineering and Mathematical Sciences, City University, London EC1V 0HB, UK.

and also to

Dr. A. Umerski.
Department of Mathematics, Open University, Milton Keynes, MK7 6AA, UK.

Three Ph. D. Positions

Computational Materials Science

Physics Department, University of Antwerp, Belgium

At the Physics Department of the University of Antwerp three PhD positions are available in the field of computational materials physics :

1. One position to study the structural and thermodynamic properties of phase change materials based on Ge-Sb-Te alloys. These materials are used for rewritable optical data storage components (”blue ray disks”). Within this project, the phase stability of the ternary Ge-Sb-Te system will be investigated using first-principles quantum mechanical calculations within the density functional theory formalism.

2. Two positions to study the optical properties of transparent conducting oxides. Using first-principles quantum mechanical calculations, the optical properties (bandgap, absorption, ...) will be investigated. These calculations will be performed in the standard density functional theory formalism as well as through many body perturbation theory (via the Bethe-Salpeter equation).

The work will be carried out in collaboration with Prof. G. Ceder from the Massachusetts Institute of Technology (MIT) (http://www.burgaz.mit.edu/) who is also a guest professor at the physics department of the University of Antwerp. Short stays with the MIT group should be expected for the first position.

The candidates should have a Master degree in physics, chemistry or applied sciences. They should have a good knowledge of solid state physics and quantum mechanics. Experience with C/C++, Fortran and/or Linux is a plus.

For more information contact

Prof. Dr. B. Partoens	Dr. D. Lamoen
tel : +32(0)32653663	tel : +32(0)32653316
email : Bart.Partoens@ua.ac.be	email : Dirk.Lamoen@ua.ac.be

http://www.cmt.ua.ac.be/

Postdoctoral Research Associate

Department of Physics, Applied Physics and Astronomy

Rensselaer Polytechnic Institute, Troy, New York, USA

The Department of Physics, Applied Physics and Astronomy at Rensselaer Polytechnic Institute, Troy, New York invites applications for the position of a postdoctoral research associate in the area of first principles simulations. The research is supported by a grant from IBM and the project will be carried out in close collaboration between Professor Saroj K Nayak at Rensselaer and Professor Wanda Andreoni at IBM Research, Zurich. The project is related to the investigation of doping of nanoscale semiconductors and close collaboration with experimental groups is expected. Candidates with strong background in solid state physics and experience in calculations of electronic structure will be given preference. The research will involve large-scale simulations and be carried out using newly established world's most powerful university based supercomputer, Computational Center for Nanotechnology Innovations (CCNI)

http://www.rpi.edu/research/ccni/index.html.

The appointment is initially for one year and can be renewed. Interested individuals should send a resume and two reference letters to Professor Saroj K Nayak at nayaks@rpi.edu. Application material accepted until positions are filled.

Rensselaer is an Equal Opportunity/Affirmative Action Employer. Women and Minorities are strongly encouraged to apply.

Postdoctoral Position

”Simulation and metadynamics applied to the structure of crystalline and amorphous clusters”

International School for Advanced Studies (SISSA), Condensed Matter Theory Sector

Trieste, Italy

The focus of research will be centered on the modelling and simulation, using metadynamics, of clusters and other systems, to explore (classical) equilibrium and non equilibrium properties.

People involved in SISSA are Erio Tosatti and Alessandro Laio.

The ideal candidate should have a good background in solid state physics or materials science, experience with classical simulations and statistical mechanics.

The position is for two years, and the yearly net salary is 18,000 or 21,500 Euros, depending on seniority.

Paper application is required. Strict deadline: 17 December 2007.

For details, please visit: `http://www.sissa.it/main/?p=A3_B4&id=304`

Scientific Information: laio@sissa.it

Administrative information: taty@sissa.it

Postdoctoral Position

”Strong correlations and ab-initio calculations in magnetic nanocontacts”

International School for Advanced Studies (SISSA), Condensed Matter Theory Sector

Trieste, Italy

The focus of research will be centered on theoretical modeling of transport, many body physics, and/or electronic properties of nanocontacts,particularly involving magnetic degrees of freedom.

The ideal candidate should have a good background in solid state physics or materials science, extensive experience with either electronic structure calculations or many-body calculations (or both).

People involved in SISSA are Erio Tosatti, Michele Fabrizio, Giuseppe E. Santoro, Andrea Dal Corso, Denis Basko.

The position is for two years, and the yearly net salary is 18,000 or 21,500 Euros, depending on seniority.

Application deadline: Dec 17 (Mon), 2007.

Paper application is required.

For details, please visit:

`http://www.sissa.it/main/?p=A3_B4&id=304`

Scientific contact: tosatti@sissa.it

Administrative contact: taty@sissa.it

Postdoctoral Position

Institut Charles Gerhardt Montpellier, France

Ab initio electrochemistry applied to fuel cell and Li batteries

Job description

A funded postdoctoral position is available starting from january 2008 at the Institut Charles Gerhardt (Montpellier, south of France) in the area of ab initio modeling of electrochemical surface reactions. The proposal deals with the reactivity of electrochemical surfaces and interfaces. We will focus on how an applied potential can modify the structure and reactivity of surface-solvent interfaces. Various complex interfaces (water/metal, solvent/Li-electrodes) of great theoretical and industrial importance will be studied. The project offers opportunities to collaborate with French centers of excellence (Ecole Normale Suprieure de Lyon (Lyon), ENPC (Paris)) and industrial companies (IFP(Lyon)) through the Sire network funded by the French ANR (national French research agency). The candidate will have access to both national French supercomputers facilities (CINES/IDRIS) and local clusters.

Qualifications

Applicants must hold Ph.D. or equivalent in physics, chemistry or materials science. Expertise in materials modeling and simulation would be strongly desired, strong background in semi-empirical/ab initio approaches would be advantageous. Aptitude to communicate effectively with co-workers and a demonstrated ability to conduct original research, are expected. The position is full-time for one year, with a possible extension to a second year. To apply, please address a detailed CV including a list of publications, and names, addresses, e-mails, and phone numbers of two or more confidential referees to:

Dr. Jean-Sébastien Filhol	(filhol@univ-montp2.fr)
Dr. Marie-Liesse Doublet	(doublet@univ-montp2.fr)

Institut Charles Gerhardt Equipe CTMM Université Montpellier
2, Place E. Bataillon 34095 Montpellier Cedex 05 France
http://www.icgm.fr and http://www.lsd.univ-montp2.fr

Ph.D. Positions

Theory and Simulation of Materials

Imperial College London, United Kingdom

Applications are invited for Ph.D. studentships starting October 2008 in the theory and simulation of materials at Imperial College London. The students will be participants in the Thomas Young Centre – London Centre for Theory and Simulation of Materials

(`http://www.thomasyoungcentre.org`),

which offers a stimulating research environment spanning the London colleges with a programme of graduate lectures, discussion meetings and seminars. Many projects are interdisciplinary and supervised by two academic staff members.

Our main research activities are in the following areas:

- Development and applications of first-principles methods for simulating at the atomic scale, including linear-scaling methods for large systems, polarisation, molecular dynamics and thermodynamics
- Development of semi-empirical methods of atomistic simulation, with applications to complex and amorphous structures, defects and interfaces in materials
- Modelling of novel nanoscale electronic and mechanical devices, e.g., electronic noses and gas sensors; thermoelectrics; thermal rectifiers, mechanical oscillators, actuators, and bearings.
- Modelling of defect behaviour in structural materials for, e.g, hydrogen fusion power plants
- Modelling of spectroscopic and spectrometric techniques (e.g., NMR, SIMS)
- Applications to nanomaterials, nanotechnology, and biomaterials

Students should have an interest in theory and simulation of materials and should expect to obtain a first class or 2i undergraduate degree in physics, materials science, chemistry, maths or a related subject. Some proficiency in a high-level programming language such as Fortran or C is desirable but not essential.

These studentships are fully funded via the EPSRC DTA award for candidates who meet the EPSRC criteria of having been been 'ordinarily resident' in the UK for the past 3 years. For nationals of EU member states who are not UK residents this funding only covers tuition-fees.

Prospective candidates are referred to

`www.imperial.ac.uk/materials/research/phdopportunities`

and are encouraged to contact members of the academic staff (listed below) for further details of specific projects.

Prof. Mike Finnis (`m.finnis@imperial.ac.uk`)
Dr. Peter Haynes (`p.haynes@imperial.ac.uk`)
Dr. Andrew Horsfield (`a.horsfield@imperial.ac.uk`)
Dr. Arash Mostofi (`a.mostofi@imperial.ac.uk`)
Dr. Paul Tangney (`p.tangney@imperial.ac.uk`)

Applications will be processed when received and it is expected that the posts will be filled by April 2008.

Postdoctoral Fellow in Ab Initio Calculations

High Temperature Materials Group

Paul Scherrer Institute, Switzerland

As part of a general program on high temperature nuclear materials development in the context of an international collaboration, the High Temperature Materials project is interested in determining the relevant parameters which affect the long term evolution of materials subjected to extreme conditions. These phenomena occur on a range of scales from the microscopic level of single crystal defects all the way up to the macroscopic continuum mechanics level. A hierarchical multi-scale modelling program to study the effects determining the lifetime of materials is currently underway. First principles ab initio calculations are at the basis of this multi-scale approach.

We are looking for a

Post doctoral fellow in ab initio calculations.

Tasks: The Post Doctoral fellow will study atomic scale defect energetics using first principles ab initio calculations in alloys using commercially available packages. The magnetic properties of the ferritic based alloys will be studied.

We are offering a two year appointment starting as soon as possible.

Candidate's profile: You have a PhD in condensed matter physics. Experience in ab initio calculations of alloys using VASP or Wien2K is required. You are able to program and run parallel codes. An understanding of magnetism is highly desirable. You must be able to communicate in English.

For further information please contact Dr Maria Samaras, e-mail maria.samaras@psi.ch.

Please send your application to: Paul Scherrer Institut, Human Resources, Mrs. Hedwig Habersaat, 5232 Villigen PSI, Switzerland, quoting

ref: 4351-01.

Scientists/Senior/Research Engineers

At Institute of High Performance Computing

Contact: HR Officer

E-mail: recruitment@ihpc.a-star.edu.sg

Address: 1 Science Park Road 1-01

The Capricorn Science Park II Singapore 117528

Please quote the job reference number when submitting the application

Simulations Of Electronic Transport In Nanodevices (IHPC/R/271107/05)

Job description: Your job scope involves, but not limited to, design and development of numerical simulations to study and predict electronics transport and other behaviour of nanodevices. You will contribute to the development of next generation nanodevices including nanowire field effect transistors using state-of-the-art high performance computing facilities and tools. The programme offers opportunities to interact with other leading international groups in nanodevices fabrications and simulations.

Requirements

A PhD/ Masters in Physics/ Applied Mathematics/ Electrical/ Electronics Engineering or its equivalent. Working/research experience in C/C++ and Fortran computing is essential Those with working knowledge and experience in nanodevice physics, semiconductors, quantum transport will have an advantage Fresh graduates with a strong interest in research and willingness to learn both numerical methodologies and engineering applications are welcome to apply

Designer Materials (IHPC/R/271107/06)

Job description: You will predict, explore and understand the fundamental properties and structure of materials as well as develop new materials using novel computational approaches such as molecular dynamics, abinitio and multi-scale methods. You will have access to state-of-the-art computing facilities and tools to study a wide range of challenging problems from green energy to electronics.

Requirements

A PhD in Physics/ Chemistry/ Mathematics/ Material Science or its equivalent. Strong interest and experience in modelling and simulation work

Multiscale Modeling of Materials (IHPC/R/271107/14)

Job description: You should possess the ability to combine different techniques for multiscale modeling of materials. These techniques are based on first-principles (density-functional based) theory, tight-binding methods, force-field methods (include coarse grained potentials), phase-field methods, and hand-shake algorithms (or other novel algorithms) to enable multiscale modeling of materials. The combined techniques will be used to perform large-scale simulations of materials such as metals and semiconductors involving point, line, and 2D defects.

Requirements

A PhD in Physics/ Chemistry/ Mathematics/ Material Science or its equivalent. Working experience in Fortran90, C, Linux environment is essential. Those with good knowledge in MPI techniques for parallelization will have an advantage.

Biophysics (IHPC/R/271107/10)

Job description: You will be expected to contribute to the development of models of the mechanical processes of cancer progression. The current areas of research include modelling the deformation and migration of cells during metastasis, molecular dynamic simulations of integrin receptors and focal adhesions, level-set models of tumor growth, etc.

Requirements

A PhD in Computational Physics, Applied Mathematics, Bioengineering, Molecular and Cell Biology, or its equivalent Working knowledge of molecular and cell biology Background in mathematical modelling and programming (C++ or Fortran)

Computational Fluid Mechanics (IHPC/R/271107/11)

Job description: You will be involved in research and applications of computational fluid mechanics. The areas of research include multiphase and interfacial flow, micro/nanofludics and fluid-structure interactions.

Requirements

A PhD in Fluid Mechanics, Physics, Applied Mathematics or its equivalent Preferably with experience in the development and application of CFD codes in a research organisation, university or industry Candidates who are capable of acquiring and managing research and industrial projects will have an added advantage

Heterogeneous Coupled System (IHPC/R/271107/13)

Job description: You will be involved in developing computational models to enable the investigation of the physical processes involved in nucleation and crystal growth. You should have a general interest in science and be able to apply your knowledge/skills to a variety of problems.

Requirements

A PhD in Physics, Mathematics, Chemical Engineering, Mechanical Engineering, Material Science or its equivalent Experience in the development of computational algorithms (C/C++, Fortran) and use of popular commercial engineering software Knowledge of molecular dynamics simulation and/or computational thermodynamics is desirable

Postdoctoral Position

Computational Spectroscopy at the Nanoscale
SISSA, Trieste, Italy

A postdoctoral position in Computational spectroscopy at the nanoscale is available at SISSA, to work with Stefano Baroni. This position is part of a theoretical effort, coordinated by the DEMOCRITOS National Simulation Center in Trieste, to develop and implement new methods that will extend the scope of ab initio calculations, based on time-dependent density-functional theory and many-body perturbation theory, to systems of hundreds of atoms. The postdoctoral fellow will concentrate on TDDFT and will have the task to implement our newly developed Liouville-Lanczos scheme in a PAW environment, and/or to develop new methods to simulate optical dichroism in extended systems, and/or electron energy-loss spectra, to name a few possible applications.

The successful candidate will be part of a strong theoretical group at SISSA and DEMOCRITOS and will have the chance to collaborate with experimentalists at the ELETTRA synchrotron radiation laboratory in Trieste.

We are seeking highly qualified candidates with a strong inclination towards methodological innovation, but equally versed in applications and able to speak to and to understand experimentalists.

Trieste is a beautifully located medium-sized city on the side of the Adriatic sea. The quality of life in the Trieste area is consistently praised in the reports of past and present PhD students and postdoctoral fellows. Local social life is lively, with cultural and natural attractions (Venice, the Alps, and beautiful seaside resorts) less than two hours away from the center of the city. Although the knowledge of the Italian language will of course facilitate social life, this is not strictly required for professional purposes, as English is the language currently spoken at SISSA. The salary being offered is compatible with a quite decent living standard.

The deadline for submitting an application is end of January 2008. Perspective candidates are invited to ask more information and to send their CV to Stefano Baroni, baroni@sissa.it. The formal announcement of the vacancy, with instructions on how to apply can be found at:

`http://www.sissa.it/main/?p=A3_B4&what=tpd`.

Postdoctoral Position
University of California, Santa Barbara, USA

A postdoctoral position in Computational Materials Physics is available to work with Prof. Chris Van de Walle in the Materials Department and the Solid State Lighting and Energy Center at the University of California, Santa Barbara (UCSB). Candidates should have experience with electronic structure and condensed matter theory. They should also have a keen interest in materials physics and a motivation to interact with experimentalists. Good communication skills (writing papers, making presentations) are required.

The project will encompass one or more of the following areas: first-principles calculations (density-functional theory) for surfaces, interfaces, and point defects; development/application of techniques to overcome band-gap problems; and modeling of device structures, including Schroedinger-Poisson simulations and/or k.p calculations of band structure.

Applications should include: (1) Curriculum Vitae, (2) Publication List, (3) One-page summary of accomplishments to date, and their significance, (4) Names and contact information (including email) of three references.

Professor Van de Walle's group works on a variety of topics ranging from new materials for solid-state light emitters to hydrogen-storage materials. The Solid State Lighting and Energy Center, directed by Profs. S. Nakamura and S. DenBaars, comprises more than 50 researchers, including faculty, visitors, postdocs and graduate students, and is focused on developing novel technologies for solid-state light emitters and energy conversion. The UCSB campus offers a unique environment, both in terms of collaborative research and natural beauty. Competitive salaries are offered, based on qualifications and experience.

More information about UCSB, about the Solid State Lighting and Energy Center, and about the Materials Department can be found at

http://www.ucsb.edu, http://ssldc.ucsb.edu, and http://www.materials.ucsb.edu.

Information about Prof. Van de Walle's research group can be found at

`http://www.mrl.ucsb.edu/~vandewalle`.

For more information or to apply, please contact Prof. Chris Van de Walle (vandewalle@mrl.ucsb.edu).

Chris G. Van de Walle
Professor
Materials Department
University of California
Santa Barbara, CA 93106-5050

Ph. (805) 893-7144
Fax (805) 893-8983
e-mail: vandewalle@mrl.ucsb.edu

POSTDOCTORAL POSITION IN COMPUTATIONAL MATERIALS SCIENCE

Uppsala University, Sweden

A postdoctoral position is available in the Theoretical Magnetism Group, Department of Physics, Uppsala University, Sweden.

The research focusses on issues related to the electronic structure of bio-materials. To be investigated are novel functional properties of large bio-molecules, applying ab initio Density Functional Theory-based techniques as well as beyond DFT approaches. Theoretical modeling of optical, magnetic, and transport properties will be performed.

Prospective candidates are expected to hold a PhD and have an excellent background in theoretical or computational physics or chemistry. Experience with large scale ab initio calculations is desirable, as is also the ability to collaborate in a multidisciplinary environment.

The Physics Department of Uppsala University offers a very strong research environment for computational material science and condensed matter theory, and it provides an excellent scientific working place for scientists.

The postdoctoral position is initial for one year; a continuation with one more year can be considered at the end of the first year.

Qualified candidates are invited to send a CV, list of publications, and names of two reference persons, to the address given below (deadline December 20, 2007).

Peter Oppeneer
Assoc. Professor, Dept. of Physics
Uppsala University, Box 530
S-751 21 Uppsala, Swede
peter.oppeneer@fysik.uu.se

Postdoctoral Position in Computational Materials Science

University of Vienna
Faculty of physics
http://cms.mpi.univie.ac.at

A postdoctoral position is available in the area of Computational Surface Science, starting in February 2008. It is associated with the group *Computational Materials Physics* at the Faculty of Physics, University of Vienna. The Group has developed and maintains the Vienna ab initio package (**VASP**), which is used world wide by nearly 1000 academic and industrial groups. An outstanding environment for acquiring first hand knowledge on density functional computations and program development is offered.

The project is focused on the development of new nano-structured surfaces using metallic or oxidic supports with the help of density functional theory calculations. The research will be performed within the framework of a national joint research program, involving both experimental and computational groups.

The candidate should be familiar with density functional theory methods and have a profound knowledge of surface science and solid state physics. Experience with kinetic Monte-Carlo simulations would be an advantage.

Interested applicants should send (1) a CV including a publication list, (2) one or two reprints representative of previous research, and (3) two confidential letters of recommendation or contact information for references to:

Dr. Florian Mittendorfer
Facultiy of Physics
University of Vienna
Sensengasse 8/12
A-1090 Wien, AUSTRIA

or

Florian.Mittendorfer@univie.ac.at

When sent by email, the application should include the CV and reprints as pdf files as well as a short cover letter in the email body.

Consideration of candidates will begin immedieatly and will continue until the positions are filled.

6 Abstracts

LDA+Gutzwiller Method for Correlated Electron Systems

XiaoYu Deng[1] , Xi Dai[1] , Zhong Fang[1,2]
[1] *Beijing National Laboratory
for Condensed Matter Physics and Institute of Physics,
Chinese Academy of Sciences, Beijing 100080, China*
[2] *International Center for Quantum Structure,
Chinese Academy of Sciences, Beijing, 100080, China*

Abstract

Combining the density functional theory (DFT) and the Gutzwiller variational approach, a LDA+Gutzwiller method is developed to treat the correlated electron systems from ab-initio. All variational parameters are self-consistently determined from total energy minimization. The method is computationally cheaper, yet the quasi-particle spectrum is well described through kinetic energy renormalization. It can be applied equally to the systems from weakly correlated metals to strongly correlated insulators. The calculated results for SrVO3 , Fe, Ni and NiO, show dramatic improvement over LDA and LDA+U

(Submitted to Phys. Rev. Let)
The REVTEX version of this paper is available on http://arxiv.org/abs/0707.4606 Correspondence: zfang@aphy.iphy.ac.cn

Interstitials in FeCr alloys studied by density functional theory

T. P. C. Klaver[1], P. Olsson[2], and M. W. Finnis[3]
[1]*School of Mathematics and Physics, Queen's University Belfast,*
Belfast BT7 1NN, Northern Ireland, UK
[2]*Department of Materials and Mechanics of Components, EDF-R&D,*
Les Renardirés, F-77250 Moret-sur-Loing, France
[3]*Department of Materials, Imperial College London,*
Exhibition Road, London SW7 2AZ, UK

Abstract

Density Functional Theory calculations have been used to study relaxed interstitial configurations in FeCr alloys. The ionic and electronic ground state of 69 interstitial structures has been determined. Interstitials were placed in alloys with up to 14 atom percent Cr. Cr atoms were either monatomically dispersed or clustered together within a periodically repeated supercell consisting of 4 x 4 x 4 cubes of bcc unit cells. The distance between the interstitials and Cr atoms was varied within the supercells. It is shown that Cr atoms beyond third nearest neighbour distance from the interstitial can still have an interaction with it of up to 0.9 eV. The multi-body nature of the Cr-Cr interactions causes the Cr-interstitial interaction to be strongly concentration-dependent. The Cr-Cr interaction in defect-free alloys is also dependent on the overall Cr concentration. The effective Cr-Cr repulsion is weaker in alloys than in an environment of pure Fe. Apart from the Cr concentration the Cr-interstitial interaction also depends on the dispersion level of Cr atoms beyond third nearest neighbour distance from the interstitial. The formation energy differences between dumbbell interstitials with different orientations are independent of the Cr concentration. We show that the long-range influence of Cr atoms on the interstitial is not due to the interstitial strain field protruding into Cr-rich parts of the supercells. The Fermi-level and band energies were found not be the sole governing parameter in determining the formation energies. Implications for the construction of empirical potentials are discussed.

(Approved for publication in Phys. Rev. B)
Contact person: t.p.c.klaver@qub.ac.uk

Structural Properties and Magic Structures in Hydrogenated Finite and Infinite Silicon Nanowires

A. D. Zdetsis*, E. N. Koukaras and C. S. Garoufalis
Department of Physics, University of Patras, Patra, GR-26500 Greece

Abstract

Unusual effects such as bending and ”canting”, related with the stability have been identified by *ab initio* real-space calculations for hydrogenated silicon nanowires. We have examined in detail the electronic and structural properties of finite and infinite nanowires as a function of length (and width) and have developed stability and bending rules, demonstrating that ”magic” wires do not bend. Reconstructed 2x1 nanowires are practically as stable as the ”magic” ones. Our calculations are in good agreement with the experimental data of Ma *et al.* [*Science* **299**, 1874 (2003)].

(Submitted to Applied Physics Letters **91**, 203112, 2007)
Contact person: zdetsis@upatras.gr

High-symmetry high-stability silicon fullerenes: A first-principles study

Aristides D Zdetsis
Department of Physics , University of Patras, Patra, GR 26500, Greece

Abstract

It is shown by *ab* initio calculations that very stable high-symmetry fullerenes of the form Si_nH_n, n=20, 28, 30, 36, 50, and 60, can be formed with large energy gaps suitable for optoelectronic and other applications. Quantum confinement seems to be violated if one neglects the essentially two-dimensional nature of the electron gas. Comparison with similar carbon fullerenes, such as $C_{20}H_{20}$ and $C_{60}H_{60}$, reveals full similarity in their electronic and geometrical structures, which is suggestive of possible routes for their synthesis. These silicon fullerenes constitute the best manifestation of carbon and silicon homology.

(Submitted to Physical Review B **76**, 075402 (2007)
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Stabilization of flat aromatic Si_6 rings analogous to benzene: *Ab initio* theoretical prediction

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Abstract

It is shown by *ab initio* calculations, based on density functional (DFT/B3LYP), and high level coupled-cluster (CCSD(T)) and quadratic CI (QCISD(T)) methods, that flat aromatic silicon structures analogous to benzene (C_6H_6) can be stabilized in the presence of lithium. The resulting planar Si_6Li_6 structure is both stable and aromatic, sharing many key characteristics with benzene. To facilitate possible synthesis and characterization of these species, routs of formation with high exothermicity are suggested and several spectral properties (including optical absorption, infrared and Raman) are calculated.

(Submitted to Journal of Chemical Physics - 2007)
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Proton exchange of small hydrocarbons over acidic chabazite: Ab-initio study of entropic effects.

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Abstract

The proton exchange reaction of a series of short hydrocarbons over an acidic zeolite (chabazite) has been studied by means of periodic DFT calculations. We show that the chain length of hydrocarbons does not have a significant effect on the height of the potential energy barrier. The experimentally observed regioselectivity between methyl and methylene groups in propane and between methyl and methine groups in isobutane is shown to be an entropic effect. In addition to the direct H-exchange, a mechanism mediated by a methylpropene molecule recently suggested by experimentalists has been explored. Again we find that entropy plays a very important role in driving the reaction.

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Charge Transfer and Polarization of Rh/MgO(001)

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Abstract

Spin-polarised plane-wave density functional calculations (using GGA-PW91) are presented for the Rh/MgO(001) interface, with coverages from 1/8 ML to 1 ML. Three stable adsorption sites are found, with that above O always the most stable. Adhesion energies lie in the range 0.6-2.1 eV for low coverage, decreasing to 0.1-1.0 eV at full coverage. Adsorbed Rh remains magnetic for all cases except 1 ML at the O site. Upon adhesion we find a reversal of the weak surface rumpling: O atoms protrude for clean MgO(001), but Mg atoms protrude for Rh/MgO(001). Projected densities of states and maps of charge density differences and the electron localization function show that the effect of Rh on the slab is shallow and rather local, with significant effects only for the Rh and the surface layer, mostly just the closest O atom.

In all cases, we find Bader charge transfer *from* the closest surface layer O *to* the Rh, while both the Rh and the surface layer O and Mg atoms become polarized. These two effects largely account for all the electronic and structural properties of the system, including the adhesion itself. Indeed, the calculated adhesion energies show a rough linear dependence on the size of the charge transfer.

(Submitted to J. Chem. Phys.)
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Correlations between magnetic properties and bond formation in Rh-MgO(001)

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Abstract

We present the results of first principles calculations for the magnetism of Rh adlayers on MgO(001), at three different adsorption sites and three different coverages, corresponding to 1, 1/2 and 1/8 monolayers. Finite magnetization is found in all cases except that of one Rh monolayer above the oxygen site, which is also the most stable. We examine how the magnetization changes as a function of the Rh-surface distance and relate this to changes in the real-space charge density and in the density of states (DOS) as the Rh adlayer interacts with the surface. We find that increasing either the Rh-Rh interaction strength or the Rh-surface interaction strength leads to reduced magnetization, while increasing the former drives a crossover from localized (atomic) to itinerant magnetism. Neither the magnetic transition itself, nor the localized-to-itinerant magnetism crossover, is found to be directly related to the formation of Rh-surface bonds. From a practical point of view, we predict that magnetism in the Rh-MgO(001) system is most likely to be found experimentally at reduced coverages and at low temperatures.

(Appeared as Surface Science 601 (2007) 1218-1230)
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Ab initio study of ladder-type polymers: Polythiophene and polypyrrole

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Abstract

An *ab initio* study of four polymers, polythiophene, polypyrrole, ladder-type polythiophene, and ladder-type polypyrrole is presented. Upon an analysis of the variation of the band gap when comparing the non-ladder and the ladder-type polymers, a discrepancy is found between the thiophene and the pyrrole polymer families. For polythiophene, the ladder-type polymer has a larger band gap than the non-ladder polymer whereas the opposite is found for the pyrrole polymers. This difference is explained in terms of the competition between the bond length alternation and the effect of the charge density distribution in the carbon backbone.

(Accepted in Chemical Physics Letters (doi:10.1016/j.cplett.2007.11.023))
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Structural and electronic properties of ZnO nanotubes from density functional calculations

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Abstract

The structural and electronic properties of armchair and zigzag single-walled ZnO nanotubes were studied using density functional theory within the generalized gradient approximation. It was found that the strain energy required for rolling a ZnO graphitic sheet into a tube is lower than those for BN and GaN nanotubes. Both the armchair and zigzag ZnO nanotubes were found to be direct gap semiconductors with the gaps decreasing as the nanotube diameter increases. We also show that the shape of the band edges remain unaltered.

(Nanotechnology **18**, 485713 (2007))

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Graphene: Buckle or Break

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Abstract

Can two-dimensional crystals exist in our three-dimensional world and would they stay planar or become buckled? These questions has been the topic of a number of theoretical studies in the past and have received renewed interest due to the recent observations of individual layers derived from layered materials such as BN, MoS and graphite. This article reviews recent Monte Carlo simulations (Nat. Mat. **6**, 858 (2007)) that provide strong theoretical support for the experimental observations of ripples in free standing graphene sheets.

(appeard in: Nature Materials **6**, 801 (2007))
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Ice formation on kaolinite: Lattice match or amphoterism?

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Abstract

The long-standing belief that kaolinite is one of the most efficient natural ice nucleating agents because it provides a close lattice match to the basal plane of ice is called into question. Instead we show through an extensive series of first principles calculations that amphoterism is key to many of the interesting properties of kaolinite with regard to water adsorption and ice nucleation.

(submitted to: Surface Science)
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A Combined DFT/LEED-Approach for Complex Oxide Surface Structure Determination: $Fe_3O_4(001)$

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Abstract

A combination of density functional theory (DFT) calculations and low energy electron diffraction (LEED) analysis is used to determine the surface structure of $Fe_3O_4(001)$. We find that the surface is rich in oxygen and the observed $\sqrt{2}$ reconstruction is a result of a Jahn-Teller distortion as established by recent DFT-calculations [Phys. Rev. Lett. **94**, 126101 (2005)]. The corresponding Pendry reliability factor is 0.34. Furthermore we investigate the influence of the preparation conditions (temperature, oxygen pressure) on the LEED intensities of natural and synthetic samples. The electron scattering phase shifts used in the analysis of the LEED spectra are derived from two methods, one based on the DFT electron densities and another employing an overlap of atomic potentials with optimized muffin-tin radii. Both approaches lead to similar results.

(submitted to: Surf. Science, in print)

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Effect of surface nanostructure on temperature programmed reaction spectroscopy: First-principles kinetic Monte Carlo Simulations of CO oxidation at $\mathbf{RuO_2}(110)$

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Abstract

Using the catalytic CO oxidation at $RuO_2(110)$ as a showcase, we employ first-principles kinetic Monte Carlo simulations to illustrate the intricate effects on temperature programmed reaction spectroscopy data brought about by the mere correlations between the locations of the active sites at a nanostructured surface. Even in the absence of lateral interactions, this nanostructure alone can cause inhomogeneities that cannot be grasped by prevalent mean-field data analysis procedures, which thus lead to wrong conclusions on the reactivity of the different surface species.

(accepted at: Phys. Rev. Lett.)
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Exciting prospects for solids: Exact-exchange based functionals meet quasiparticle energy calculations

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Abstract

Focussing on spectroscopic aspects of semiconductors and insulators we will illustrate how quasiparticle energy calculations in the G_0W_0 approximation can be successfully combined with density-functional theory calculations in the exact-exchange optimised effective potential approach (OEPx) to achieve a first principles description of the electronic structure that overcomes the limitations of local or gradiant corrected DFT functionals (LDA and GGA).

(accepted at: physica status solidi (b))
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Consistent set of band parameters for the group-III nitrides AlN, GaN, and InN

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Abstract

We have derived consistent sets of band parameters (band gaps, crystal field-splittings, band gap deformation potentials, effective masses, Luttinger and E_{P} parameters) for AlN, GaN, and InN in the zinc-blende and wurtzite phases employing many-body perturbation theory in the G_0W_0 approximation. The G_0W_0 method has been combined with density-functional theory (DFT) calculations in the exact-exchange optimized effective potential approach (OEPx) to overcome the limitations of local-density or gradient-corrected DFT functionals (LDA and GGA). The band structures in the vicinity of the Γ-point have been used to directly parameterize a 4 x 4 $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian to capture non-parabolicities in the conduction bands and the more complex valence-band structure of the wurtzite phases. We demonstrate that the band parameters derived in this fashion are in very good agreement with the available experimental data and provide reliable predictions for all parameters which have not been determined experimentally so far.

(submitted to: Phys. Rev. B)

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7 Presenting Other Initiatives

7.1 SaX: An Open Source Package for the Calculation of Electronic and Optical Properties

SaX Developers Team and INFM-CNR S3

http://www.sax-project.org

Main developers: L. Martin-Samos and G. Bussi
Scientific board: L. Martin-Samos, G. Bussi, A. Ruini, M. J. Caldas and E. Molinari

`SaX`, which stands for Self-energy And eXcitations, is an open-source GNU General Public License package for the calculation of electronic excitations, developed at the national Research Center on nanoStructures and bioSystems at Surfaces (S3) in Modena, of the Italian INFM-CNR (`http://www.s3.infm.it`).

We are pleased to inform the psi-k community that the Release 1.0.0 is now available at `http://www.sax-project.org`.

`SaX` works in the framework of many-body perturbation theory in the GW approximation. This is the state-of-the-art method to describe electronic band structures and excitonic effects beyond the density-functional theory (DFT). The package implements the GW approximation and the Bethe-Salpeter Equation using periodic cells, plane-waves and pseudopotentials, and the various matrix elements are calculated on real-space grids through fast-Fourier transforms.

`SaX` calculations will provide the user with:

- Full dynamical structure of the random-phase-approximation polarizability.
- Quasiparticle energies and wavefunctions.
- Different implementations of GW scheme.
- Different degrees of self-consistency in the GW scheme.
- Excitonic effects with the Bethe-Salpeter equation.

`SaX` works, in principle, as postprocessing of a previous DFT calculation. In the present version, the package is interfaced through a wrapper with the DFT codes of the Quantum-Espresso distribution (`http://www.quantum-espresso.org`).

For further information about `SaX`, please, visit the web page at `http://www.sax-project.org` or contact us at saxteam@unimore.it.

8 SCIENTIFIC HIGHLIGHT OF THE MONTH: Evolutionary crystal structure prediction: overview of the USPEX method and some of its applications

Evolutionary crystal structure prediction: overview of the USPEX method and some of its applications

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Prediction of the stable crystal structure on the basis of only the chemical composition is one of the central problems of condensed matter physics, which for a long time remained unsolved. The recently developed evolutionary algorithm USPEX (Universal Structure Predictor: Evolutionary Xtallography) made an important progress in this field, enabling efficient and reliable prediction of structures with up to 30-40 atoms in the unit cell using *ab initio* methods (or up to 100-200 atoms/cell with classical forcefields). Here we review this methodology, illustrating its variation operators and visualising evolutionary runs using specifically formulated similarity matrices. We also show several recent applications – (1) prediction of new high-pressure phases of $CaCO_3$, (2) search for the structure of the polymeric phase of CO_2 ("phase V"), (3) search for new phases of transition metals, (4) high-pressure phases of oxygen, (4) new high-pressure phases of FeS and FeO, (5) exploration of possible stable compounds in the Xe-C system at high pressures, (6) investigation of previously proposed cluster-based insulators $Al_{13}K$ and $Al_{12}C$.

1 Introduction

The problem of finding the crystal structure stable at given pressure and temperature conditions, knowing just from chemical composition, occupies a central place in materials design. The ability to solve this problem would open new ways also for the understanding of the behaviour of materials at extreme conditions, e.g. those found in deep planetary interiors.

The simplest method is to explore relative energetics (i.e. compare the free energies) of a number of candidate structures (which can be known structures of analogous materials, or new structures guessed using chemical intuition), and this seems to be the most popular approach. Problems arise almost every time when a totally unexpected and hitherto unknown structure is stable. Instead of relying on real experiments and human intuition, the more advanced approach of data mining [2] applies methods from machine learning to derive rules of stability of crystal structures from a large set of *ab initio* calculations. A number of simpler intuitive empirical schemes (e.g. structure diagrams, polyhedral clusters – see [3]) have appeared in literature, but unlike the data mining approach, their application usually requires a large experimental data set or good understanding of the compound at hand.

Reliable structure prediction can be performed without any prior knowledge or assumptions about the system. Simulated annealing [4]-[7], minima hopping [8] and metadynamics [9]-[11] have been used with some success. For very small systems, even relaxing randomly produced structures can deliver the stable structure [12]. In this article, we review the evolutionary algorithm USPEX (Universal Structure Predictor: Evolutionary Xtallography) [13]-[15] and illustrate several of its applications. Section II presents basics of the method, section III shows several interesting test cases (mostly on systems with a known ground state), while a number of applications to systems where the stable structure is unknown are presented in section IV. This review contains both new results and those published previously ([13][14][30][32][69][73]).

2 Evolutionary Algorithm USPEX

Several groups attempted the pioneering use of evolutionary algorithms to structure prediction - for crystals [19]-[22], colloids [23] and clusters [24]. The algorithm developed by Deaven and Ho [24] is perhaps especially interesting as some of its features (real-space representation of structures, local optimisation and spatial heredity) are similar to the USPEX method. Their algorithm has successfully reproduced the structure of the C_{60} buckminsterfullerene, but has never been extended to heteroatomic clusters, nor to periodic systems (i.e. crystals). The algorithm of Bush and Woodley [19]-[21] was originally developed for crystals and successfully produced a starting model for solving the structure of Li_3RuO_4 [19]. However, subsequent systematic tests [20][21] showed frequent failures even for rather simple systems containing ~10 atoms/cell. Other drawbacks are that this algorithm requires experimental lattice parameters and simulations are very expensive, unless a cheap and crude heuristic expression is used for fitness. Unlike the Deaven-Ho algorithm and USPEX, in this method structures are represented by binary “0/1” strings, there is no local optimization and no spatial heredity.

In USPEX, structures are represented by fractional coordinates for the atoms and lattice vectors. USPEX operates with populations of structures; from a population, parent structures are selected. The fitness of structures is the relevant thermodynamic potential derived from *ab initio* total energy calculations. The worst structures of a population are discarded; for the remaining structures the probability of being selected as parent is a function (e.g. linear) of its fitness rank. A new candidate structure is produced from parent structures using one of three operators: (i) heredity, which combines spatially coherent slabs (in terms of fractional coordinates) of two parent structures, while the lattice vectors matrices are weighted averages of the two parent lattice vectors matrices, (ii) permutation (as in [20][21]), which swaps chemical identities in randomly chosen pairs of atoms, (iii) lattice mutation, which distorts the cell shape by applying a random symmetric strain matrix. To avoid pathological lattices, all newly produced structures are rescaled to produce a predefined unit cell volume (a reasonable starting value should be supplied in the input, and then allowed to evolve during the run). Heredity enables very broad searches, while preserving already found local fragments of good structures, and introduces ideas of "two-phase" simulations. Permutation facilitates finding the optimal ordering of the atoms; in some situations (for systems with a large range in degree of chemical similarity between different atom types) it may be useful to swap only chemically more similar atoms (e.g. Al-Si in aluminosilicates). Lattice mutation enables better exploration of the neighbourhood of parent structures, prevents premature convergence of the lattice, and essentially incorporates the ideas of metadynamics in our search. The action of these variation operators is illustrated in Figs. 1,2.

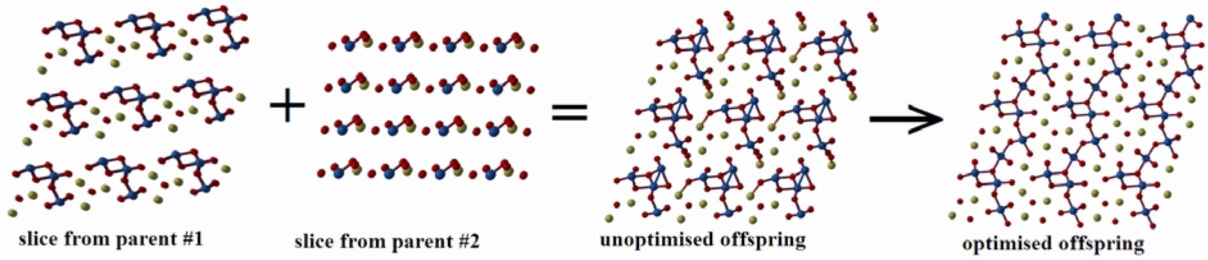


Fig. 1. Heredity operator: slices of two parent structures, and the offspring structure before and after local optimisation.

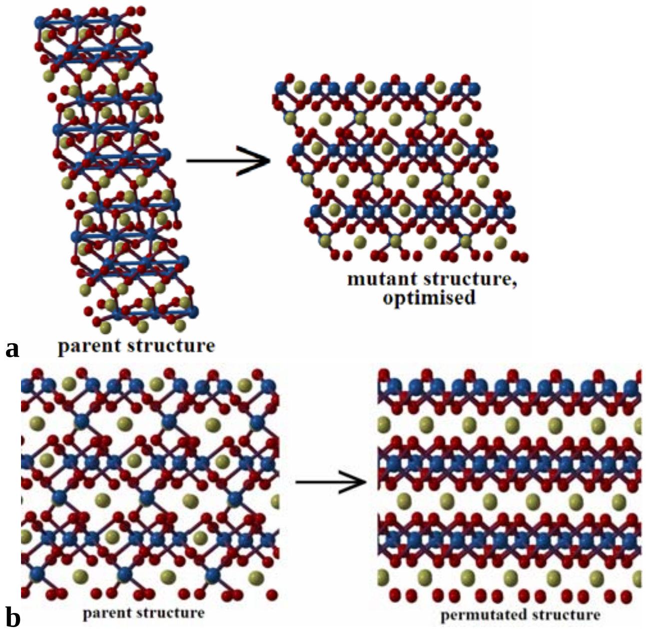


Fig. 2. Illustrations of lattice mutation and permutation operators.

Before new candidate structures are relaxed, they are tested against three constraints – first, all interatomic distances must be above the specified minimal values; second, cell angles must be between 60^{o} and 120^{o}; third, all cell lengths must be larger than a specified value (e.g. diameter of the largest atom). These constraints help to ensure stability of energy calculations and local optimization, and remove only redundant and infeasible regions of configuration space - thus the search is physically unconstrained. If in violation of these constraints, the candidate structure is discarded; otherwise, it is locally optimized (relaxed). Structure relaxations and energy calculations are done by external codes (currently, USPEX is interfaced with VASP [25], SIESTA [26], GULP [27]).

The relaxed structures are recorded and used for producing the next generation of structures. A new population of structures is made to contain one or more lowest-enthalpy structures from the previous population and the new structures produced using variation operators. The above procedure is repeated in a loop.

The first generation usually consists of random structures, but it is possible to include user-specified structures. If lattice parameters are known, runs can be done in the fixed cell, but this is not required and in most cases simulations are done with variable cell shape.

Recently [69] we improved the algorithm by more exhaustive removal of lattice redundancies. The constraint that the cell angles be between 60º and 120º (in simulations we use a slightly wider interval) removes only part of the redundancies and still allows inconvenient redundant cell shapes to be produced (e.g. cells with $\alpha=\beta=\gamma\sim120^{\circ}$ are practically flat). Certain advantages would be gained by transforming such cells to a cell shape with shorter cell vectors. Such a transformation can be done if there is at least one lattice vector whose projection onto any other cell vector (or onto the diagonal vector of the opposite face of the unit cell) is greater (by absolute value) than half the length of that vector, i.e. for vectors **a** and **b**, these criteria are:

$$\left|\frac{\mathbf{a}\bullet\mathbf{b}}{|\mathbf{b}|}\right| > \frac{|\mathbf{b}|}{2} \qquad (1a)$$

$$\left|\frac{\mathbf{a}\bullet\mathbf{b}}{|\mathbf{a}|}\right| > \frac{|\mathbf{a}|}{2} \qquad (1b)$$

E.g. for the criterion (1a) the new vector $\mathbf{a}^*$ equals:

$$\mathbf{a}^* = \mathbf{a} - ceil\left(\left|\frac{\mathbf{a}\bullet\mathbf{b}}{|\mathbf{b}|}\right|\right)(sign(\mathbf{a}\bullet\mathbf{b}))\mathbf{b} \quad , \qquad (2)$$

During this transformation atomic fractional coordinates are transformed so that the original and the transformed structures are identical (during the transformation Cartesian coordinates of the atoms remain invariant). For more details on the USPEX method, see [14][15].

An evolutionary algorithm, somewhat similar to USPEX, was proposed slightly later and independently from us in [71]; this method differs from USPEX in the absence of permutation (i.e. potential problems for binary and more complex compounds), different forms of heredity and mutation, absence of cell rescaling. Much later, our USPEX method was reimplemented in paper [72], which contained a number of errors (see comment [73]), but for very simple test cases was also successful.

Why is the USPEX methodology successful? One of the reasons is that local optimization with high probability creates chemically reasonable local environments of the atoms. Evolutionary pressure, through selection, forces the population to improve from generation to generation. In heredity, local arrangements of atoms (spatially coherent pieces of structures) are partly preserved and combined. This respects the predominant short-ranged interactions in crystals and exploits information from the current population. For large systems it may be advantageous to combine slabs of several structures. On the other hand, for systems with very few atoms (or molecules) in the unit cell heredity becomes obsolete (in the limit of 1 atom/unit cell it is completely useless); these cases, however, are trivial for other variation operators and even for local optimization of random structures.

Another important reason is that the energy landscapes expected in chemical systems are likely to have an overall "funnel" shape (Fig. 3), where lowest-energy structures are clustered in the same region of configuration (or order parameter) space. In such cases, evolutionary algorithms are particularly powerful: they "zoom in" on the most promising region of configuration space until the global minimum is found. In some cases (where dramatically different atomic arrangements are energetically competitive – e.g. complex molecular crystals where different molecules can be assembled from the same atoms) the landscape is expected to contain multiple funnels, and such systems are then particularly challenging as the algorithm may tend to get stuck in one particular funnel. To avoid this, several tools can be used – including dense random or quasirandom sampling (to cover all funnels), tabu lists or special constraint techniques (to deal with each funnel, or a group of funnels, separately).

The overall landscape shape (Fig. 3) implies that, *en route* to the global minimum some of the low-energy metastable minima can be discovered. This is important, as such phases are often interesting as well. Furthermore, metastable structures found during evolutionary simulations provide a deep insight into the structural chemistry of the studied compound. Thus, evolutionary simulations provide three major results – 1) the ground-state structure, 2) a set of low-energy metastable structures, 3) detailed information on the chemical regime of the compound.

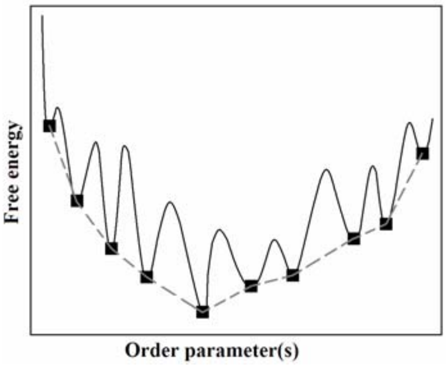


Fig. 3. Expected energy landscape in chemical systems. The original response surface is very “noisy” (i.e. contains very large energy variations, with high barriers). Local optimization reduces this surface to local minima points (black squares). The reduced response surface (dashed line) is well-behaved and has a simple overall shape. This is why the use of local optimization dramatically improves global optimization [15].

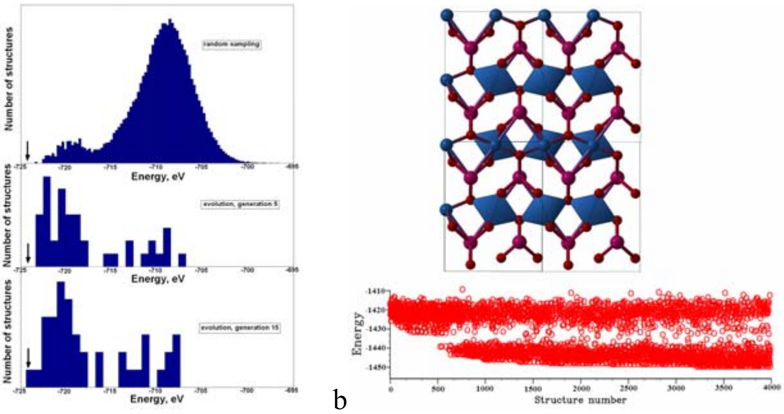
3 Tests of the Algorithm

To measure the strengths and weaknesses of the algorithm, we consider several issues:

1. efficiency of finding the global minimum, in particular relative to a simple well-defined search method, the random sampling,
2. size of systems that can be studied in practice,
3. how fast the diversity decreases along the evolutionary trajectory.

A number of successful tests have been reported in [14][15][28][69][73]. The largest successful test is for a Lennard-Jones crystal with 128 atoms in the (super)cell with variable-cell structure search, which has correctly identified hcp structure as the ground state within 3 generations (each consisting of only 10 structures). For larger Lennard-Jones systems (256 and 512 atoms/cell) we found an energetically very slightly less favourable fcc structure.

The largest test for a chemically complex system is the prediction of the structure of $MgSiO_3$ post-perovskite [33][34] using a relatively large 80-atom supercell (with fixed supercell parameters) and an empirical potential [34] describing interatomic interactions within a partially ionic model. Local optimization and energy calculations were done using the GULP code [27]. Previously [28] we have shown that already in a 40-atom supercell this test is unfeasible using the simple random sampling (with local optimization) [12]: the correct structure was not produced even after 1.2×10^5 random attempts, but was found with 600-950 local optimizations of structures produced by USPEX. With 80 atoms/cell the problem becomes much more complicated (one expects an exponential increase of complexity with system size), but even in this case we correctly produced the post-perovskite structure in a reasonable number (~3200) of local optimizations – see Fig. 4.



a

b

Fig. 4. Evolutionary prediction of the structure of $MgSiO_3$ post-perovskite using the experimental cell parameters for a) 40-atom [28] and b) 80-atom [69] supercells. In both cases, each generation consisted of 41 structures. Panel (a) compares densities of states of optimized structures generated randomly (top) and in the evolutionary run. Random sampling did not find the correct structure within 1.2×10^5 steps, whereas in the evolutionary simulation shown it was found within 15 generations (i.e. 600 local optimizations). Arrows mark the ground-state energy. Panel (b) shows the energies of structures along the evolutionary trajectory for the 80-atom run; the structure of post-perovskite was obtained within ~3200 local optimizations. One can see that the density of low-energy structures increases during the simulation.

Fig. 5 shows variable-cell *ab initio* results for $MgSiO_3$ at the pressure of 120 GPa. Several runs with somewhat different (but within a reasonable range) parameters have been performed and all produced the correct ground-state structure of post-perovskite. The number of local optimizations performed before this structure was found ranged in different runs between 120 and 390; the longest run is shown in Fig. 5. The similarity matrix (Fig. 5b) shows a slow decrease of population diversity. This is what we usually find in simulations, except for very small or very special systems. The construction of the similarity matrix is outlined in the Appendix.

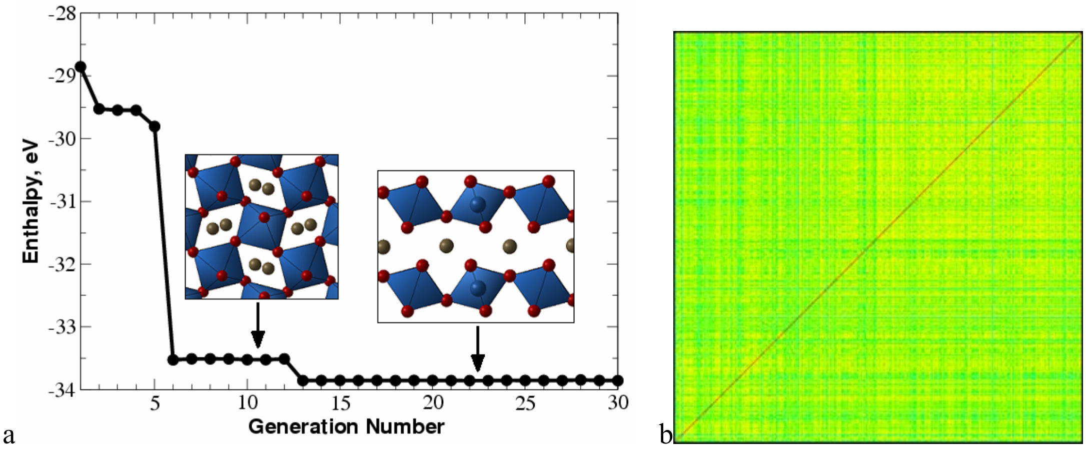


Fig. 5. Evolutionary structure search for $MgSiO_3$ at 120 GPa. a) evolution of the lowest enthalpy as a function of the generation (insets show the structures of perovskite and post-perovskite phases), from [14]. b) structure similarity matrix (dimensions 500x500).

An example of a very simple test, variable-cell *ab initio* structure search for GaAs with 8 atoms/cell [73], is given in Fig. 6. The ground-state structure for systems of such size can be found even by local optimisation of a reasonable number of randomly produced structures. The density of states of relaxed random structures (Fig. 6a), obtained from 3000 random structures, has a characteristic multimodal shape, which seems to be a general feature of energy landscapes. The stable zincblende structure has the abundance of ~0.2%, i.e. finding it with random search would on average take ~ 500 local optimizations. In evolutionary simulations (Fig. 6b) it can be found within 3 generations, or just 30 structure relaxations. Similarity matrices for random (Fig. 6c) and evolutionary (Fig. 6c) searches clearly reveal a strong increase of structure similarity (i.e. decrease of diversity) along the evolutionary run, after finding the global minimum. Even in this extreme case a significant number of dissimilar structures are produced long after the global minimum is found.

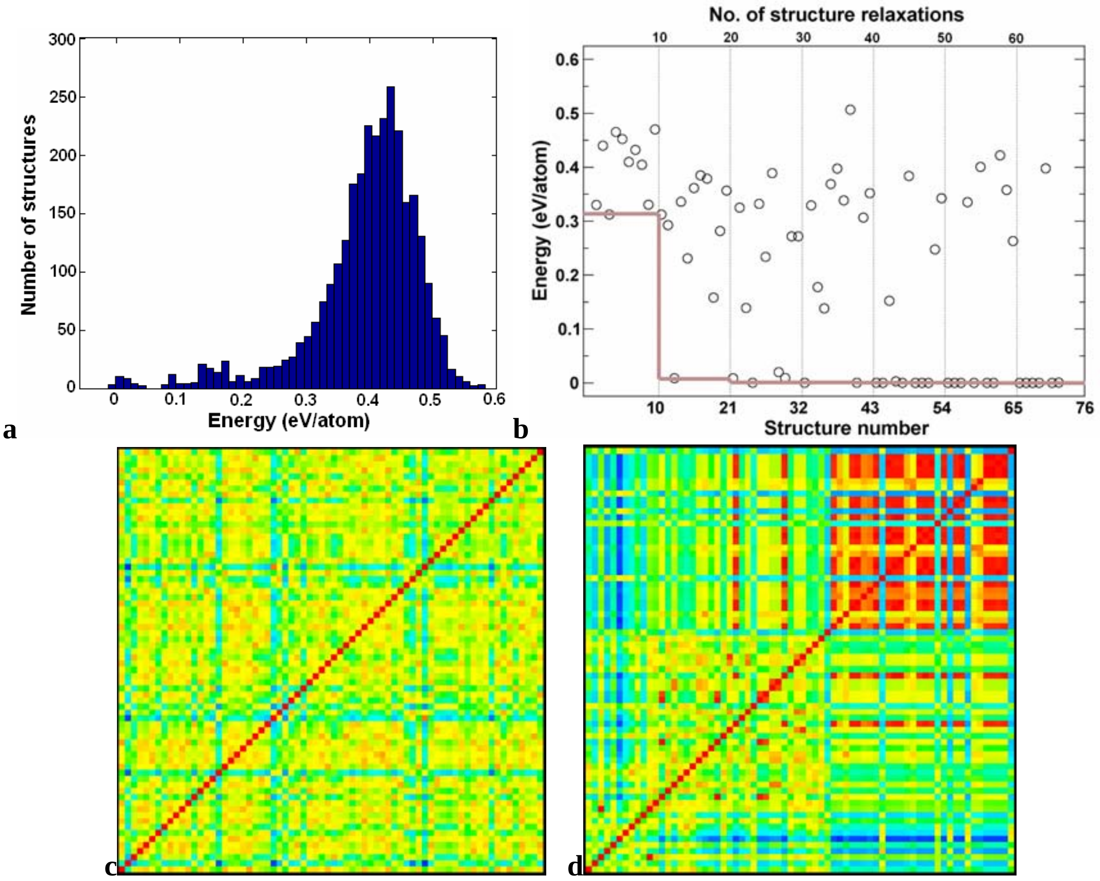
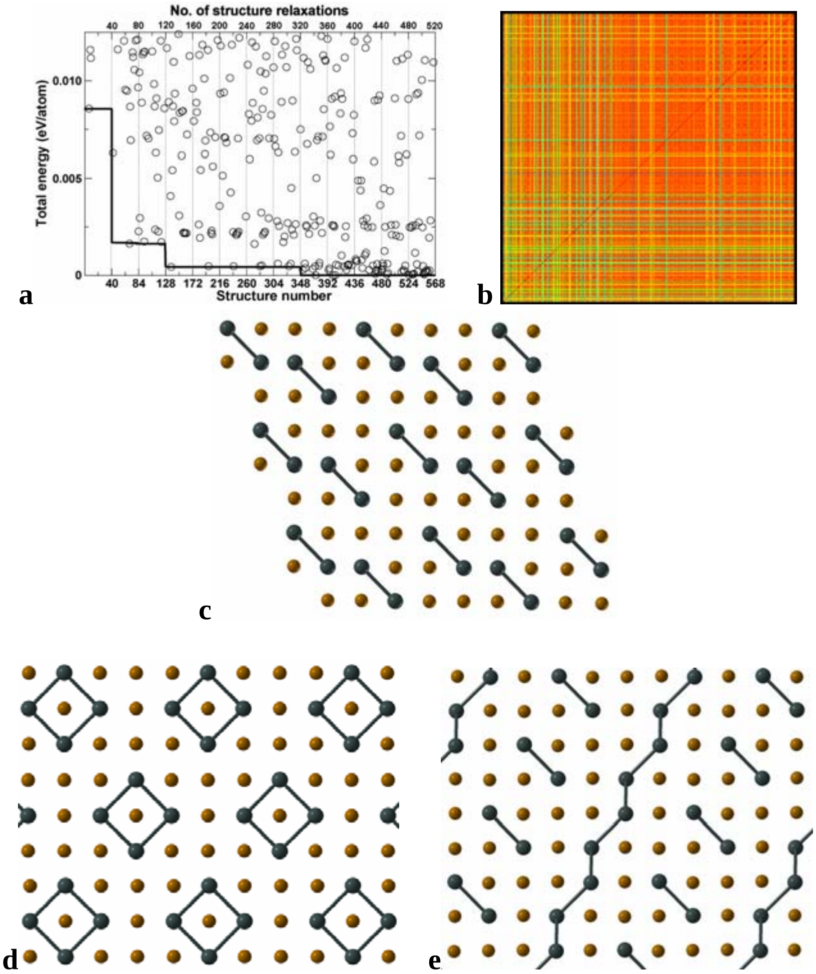


Fig. 6. Structure prediction for GaAs (8 atoms/cell): a) energy distribution for relaxed random structures, b) progress of an evolutionary simulation (thin vertical lines show generations of structures, and the grey line shows the lowest energy as a function of generation), c-d) similarity matrices (dimension 70x70) for the random and evolutionary searches, respectively. All energies are relative to the ground-state structure. The evolutionary simulation used a population of 10 structures. Panels a,b are from [73]. Calculations are performed within the GGA [38].

Au_8Pd_4 (12 atoms/cell) is an unusual system, where a number of different ordered decorations of the fcc structure have competitive energies. The ground state of this system is unknown, but was investigated in several computational studies [78][79][74][72][73]. Assuming that the ground-state structure should be an ordered variant of the cubic close-packed ("fcc") structure and using the cluster expansion technique with parameters calibrated on a set of *ab initio* energies, Barabash et al.[74] found two energetically nearly degenerate structures (Fig. 7d,e), which seemed to be the only ground-state solutions. Evolutionary search done in Ref. [72] found another fcc-based structure (Fig. 7f), which is 2 meV/atom above the ground state (4 meV/atom from more precise calculations [73]). This failure has led the authors of Ref. [72] to incorrectly conclude that evolutionary structure prediction is “not able to mimic for metal alloys the resolution of cluster expansion”. Errors that have led the authors of Ref. [72] to this conclusion have been analysed in [73]. Our evolutionary simulations [73], performed within the same approximations (the same LDA functional and ultrasoft pseudopotentials) found a new ground-state structure (Fig. 7c) that has been overlooked by the previous cluster-expansion study [74] and turned out to be ~0.1 meV/atom lower in energy than the previously known lowest-energy structures (Fig. 7d,e). Examination of all the produced structures shows that most of them are different ordering schemes of the fcc-structure and the energy differences are generally very small (Fig. 7a). The problem of structure prediction in this case reduces to that of finding the optimal ordering scheme within the basic fcc-structure; the ordering scheme is strongly coupled to the unit cell shape (especially for such small systems). Thus, maintaining a large diversity of cell shapes within a run is important and probably difficult to achieve with the small population size chosen in Ref. [72]. Similarity matrix (Fig. 7b) shows that even with these settings, the diversity in the system is rather limited and visibly decreases during the run. Reasonable choice of simulation parameters is essential for the success of any simulations; analyzing the evolutionary trajectories, the energies (e.g. Fig. 7a) and similarity matrices (Fig. 7b) for preliminary calculations can help in choosing the parameters.



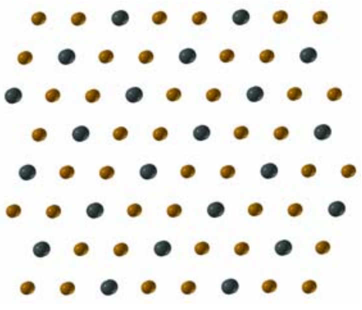


Fig. 7. Evolutionary structure search for Au_8Pd_4. a) evolution of the total energies (only the lowest-energy part is shown for clarity), b) structure similarity matrix (dimensions 873x873), c) the lowest-energy structure found in our evolutionary simulation, d-e) – the lowest-energy structures found by cluster expansion in Ref. [74] (structures No. 4905 and No. 4557, respectively, in the search catalogue of Ref. [74]), f) the lowest-energy structure found in Ref. [71]. All energies are given relative to the ground state.

4 Some Applications of the Method

In this section we will review some new insight that has been obtained using our method (see also [14]). All structure predictions described here were performed within the generalized gradient approximation (GGA [38]) and the PAW method [39][40], using VASP [25] for local optimization and total energy calculations. The predicted structures correspond to the global minimum of the approximate free energy surface. For systems where the chosen level of approximation (GGA in cases considered below) is adequate, this corresponds to the experimentally observed structure. Where this is not the case, results of global optimization are invaluable for appraising the accuracy of the approximations and may sometimes still be qualitatively correct. Most of the cases considered below (except ε-O_8, FeO and to some extent FeS) are well described by the GGA.

$CaCO_3$ polymorphs. High-pressure behaviour of carbonates is very important for the global geochemical carbon cycle, as high-pressure carbonates of Mg and Ca are expected to contain most of the Earth's carbon [43]. For $CaCO_3$, there is a well-known transition from calcite to aragonite at ~2 GPa, followed by a transition to a post-aragonite phase at ~40 GPa [45], the structure of which was solved [13] using USPEX, and the predicted structure matched the experimental X-ray diffraction pattern well. Furthermore, we have predicted [13] that above 137 GPa a new phase, with space group $C222_1$ and containing chains of carbonate tetrahedra, becomes stable. Recently this prediction was verified by experiments [46] at pressures above 130 GPa. We note that both post-aragonite and the $C222_1$ structure (Fig. 8) belong to new structure types and could not have been found by analogy with any known structures.

The presence of tetrahedral carbonate-ions at very high pressures invites an analogy with silicates, but the analogy is limited. In silicates, the intertetrahedral angle Si-O-Si is extremely flexible [48], which is one of the reasons for the enormous diversity of silicate structure types.

Fig 9 shows the variation of the energy as a function of the Si-O-Si angle in the model $H_6Si_2O_7$ molecule – method borrowed from [48]. One can see only a shallow minimum at $\angle(\mathrm{Si-O-Si})=135^{\circ}$, but a deep minimum at $\angle(C-\mathrm{O}-C)=124^{\circ}$ with steep energy variations for $H_6C_2O_6$ (Fig. 9). This suggests a much more limited structural variety of metacarbonates, compared to silicates. In both $CaCO_3$ and CO_2 the $\angle(C-\mathrm{O}-C)$ angles are close to 124° in a wide pressure range.

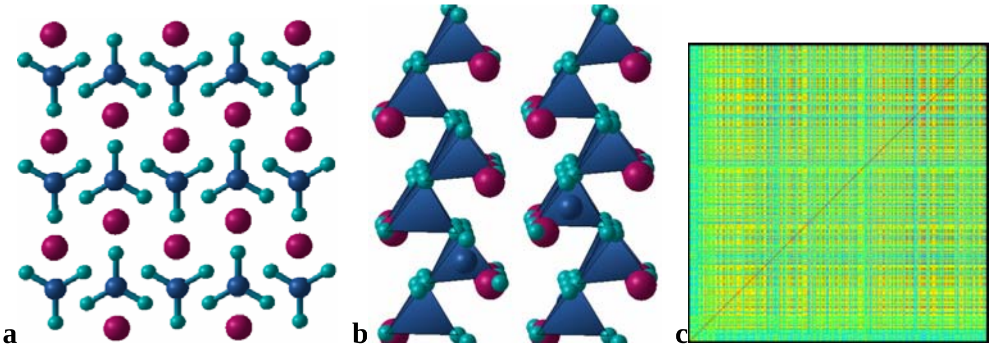


Fig. 8. $CaCO_3$ at high pressure. a) structure of post-aragonite phase, b) $C222_1$ phase, c) similarity matrix (dimensions 500x500) for $CaCO_3$ with 10 atoms/cell at 80 GPa, showing considerable diversity remaining in the population throughout the run.

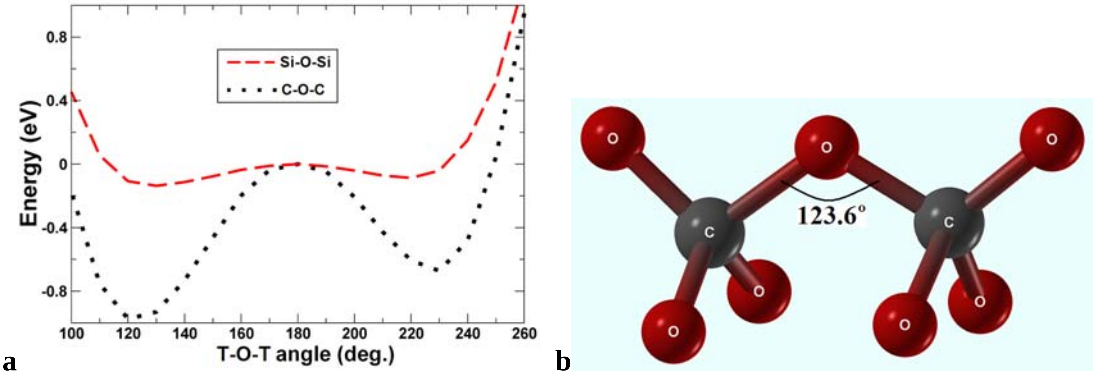


Fig. 9. Energy variation as a function of the T-O-T angle (red dashed line – T = Si, black dotted line – T = C). Calculations were performed on $H_6T_2O_7$ molecules; at each angle all T-O distances and O-T-O valence angles were optimised. Optimum angle C-O-C=124°, Si-O-Si=135°. These calculations were performed with SIESTA code [26] using the GGA functional [38], norm-conserving pseudopotentials and a double-ζ basis set with a single polarisation function for each atom.

Polymeric phase of CO_2 [50]. High-pressure behaviour of CO_2 is still controversial [51]. It is known that above ~20 GPa a non-molecular phase (called phase V [52]) with tetrahedrally coordinated carbon atoms becomes stable, but its structure is still under debate: in the first experimental study [52] a trydimite structure was proposed, but later theoretical works found it to be unstable (even not metastable) and much less favourable than the β-cristobalite structure [53][54]. At the same time, it was not possible to rule out that there may be even more stable structures. Here, we have performed evolutionary structure searches at 50 GPa, 100 GPa and 150 GPa for system with 6, 9, 12, 18 and 24 atoms/cell. At all these pressures we confirmed stability of the β-cristobalite structure (Fig. 10), thus suggesting an experimental re-investigation of phase V of carbon dioxide. CO_2-V is stable against decomposition into diamond and oxygen (the enthalpy of decomposition is very large and increases from 3.3 eV to 3.8 eV between 50 GPa and 200 GPa). Evolutionary simulations also uncovered several interesting metastable polymeric forms shown in Fig. 11. In agreement with our expectations, none of these structures are known for SiO_2. One can notice that for CO_2, the β-cristobalite structure is much lower in enthalpy compared to other structures – this is again in contrast with silica, where a large number of energetically similar structures are known. To a large extent this difference is explained by the different flexibilities of the intertetrahedral $\angle(\mathrm{T}-\mathrm{O}-\mathrm{T})$ angles (see above).

At lower pressures, between 8.9 GPa and 18.9 GPa, the $P\frac{4_2}{m}nm$ phase (see [68] for details) is stable, and at even lower pressures (0-8.9 GPa) the $Pa3$ structure is stable (Fig. 10). The $Pa3$-$P\frac{4_2}{m}nm$ transition pressure calculated here (8.9 GPa) is consistent with experiment and previous calculation [68].

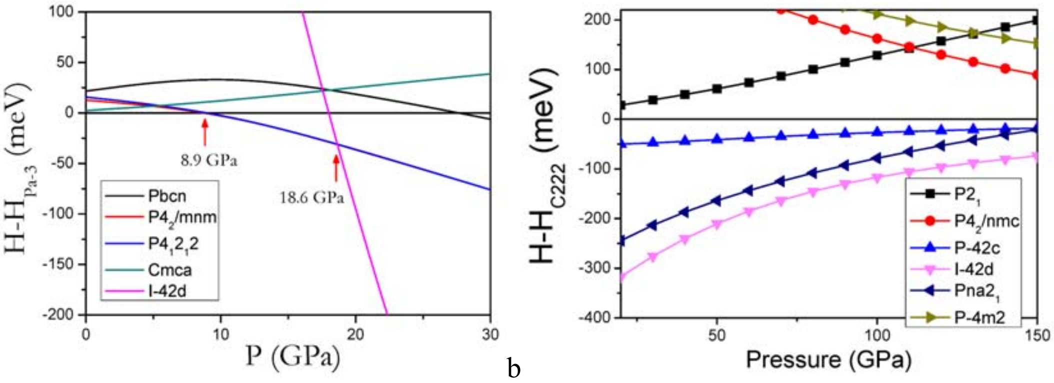
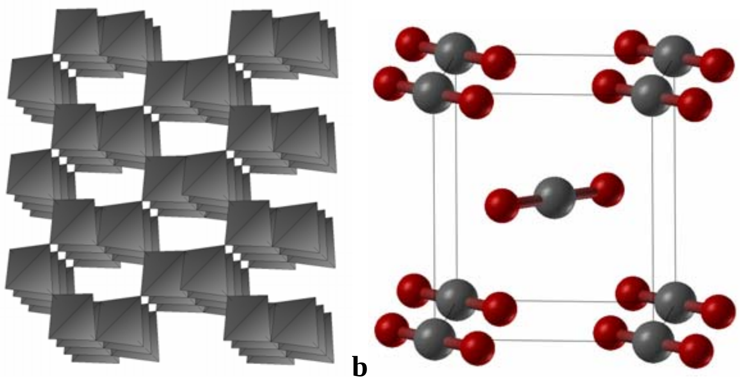


Fig. 10. Enthalpies of candidate forms of CO_2: a) in the low-pressure region, relative to the molecular $Pa3$ structure, b) in the high-pressure region, relative to the non-molecular $C222$ structure. From [50].



a **b**

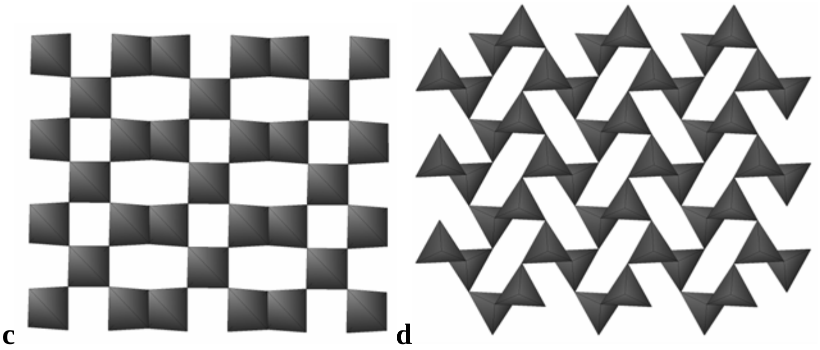


Fig. 11. CO_2 structures: a) β-cristobalite-type form of CO_2 at 100 GPa, suggested to be the structure of phase V and showing carbonate tetrahedra. Structural parameters at 100 GPa: space group $I\bar{4}2d$, a=b=3.2906 Å, c=6.0349 Å, C(0.5; 0; 0.25), O(0.2739; 0.25; 0.125), b) molecular $P\frac{4_2}{m}nm$ structure, stable at lower pressures than CO_2-V, c) polymeric $C222$ structure, d) metastable polymeric $Pna2_1$ structure. From [50].

Methane (CH_4) under pressure: formation of diamond or polymerization? Methane, one of the major components of giant planets Uranus and Neptune, is expected to become unstable at high pressure. However, neither the minimum pressure of its instability nor the product of its chemical transformation are well constrained. Some studies [80][81][82] suggested decomposition into diamond and hydrogen, others [83] proposed polymerization into heavier hydrocarbons. These problems still remain open. The simulations we have done up to now (at 100 GPa, 200 GPa, 300 GPa with 10, 15 and 20 atoms/cell in each case) are only preliminary and do not fully resolve them, but they already shed some light onto the nature of the chemical transformation.

Fig. 12a shows the most stable structure made of CH_4 molecules, which we found at 100 GPa and 200 GPa. At 200 GPa and 300 GPa we found more stable structures made of higher hydrocarbons (propane C_3H_8, butane C_4H_{10} and infinite-chain $(CH_2)_\infty$ polymer) mixed with H_2 molecules (Fig. 12b-d). To understand the chemical transformation of methane under pressure, we consider the following reactions at 300 GPa:

$$CH_4 = C + 2H_2 \quad , \qquad \text{(R1)}$$

i.e. decomposition of methane into diamond and hydrogen (structure of hydrogen was taken from our previous results [14]).

$$4CH_4 = C_4H_{10} + 3H_2 \quad , \qquad \text{(R2)}$$

i.e. decomposition into butane and hydrogen (the structure of butane was taken, as the first approximation, from the structure shown in Fig. 12d, removing the H_2 molecules)

$$2CH_4 = C_2H_4 + 2H_2 \quad , \qquad \text{(R3)}$$

i.e. decomposition into an infinite zigzag polymeric chain $(CH_2)_\infty$ structure (which here was taken, as the first approximation, from the structure shown in Fig. 12c, removing the H_2 molecules)

Additionally, we looked at the enthalpy of decomposition of butane into diamond and hydrogen (we find it to be strongly positive, i.e. butane is stable against this decomposition) and the enthalpy of removal of the H_2 molecules from the structure shown in Fig. 12d (we find it to be negative, i.e. phase separation is favourable; this also shows that the structure of butane we assumed here is energetically at least very reasonable).

At 300 GPa, we find reaction (R1) to be unfavourable (by 0.04 eV); since its products are 4% denser than reactants, it will become favourable at higher pressures. Reaction R2, however, is extremely favourable (by 0.72 eV) already at 300 GPa; its products are also significantly (2%) denser than the reactants and this reaction should be preferred to R1 up to very high pressures.

How far does polymerization go under pressure? Decomposition into diamond and hydrogen (R1) is unstable, and so is the formation of infinite polymers (R3, unfavourable by 0.29 eV). Therefore, we expect the formation of finite alkane polymers, such as butane, to be the most favourable process in a wide pressure range. It is possible that with pressure the length of the polymers would increase (a coexistence of different polymers is likely in the compressed fluid [83], and a gradual increase in fraction of heavier polymers is possible on increasing pressure). To resolve these issues and obtain precise polymerization pressure for methane, we need to perform additional evolutionary simulations for other compositions (C_3H_8, C_4H_{10}, etc.)

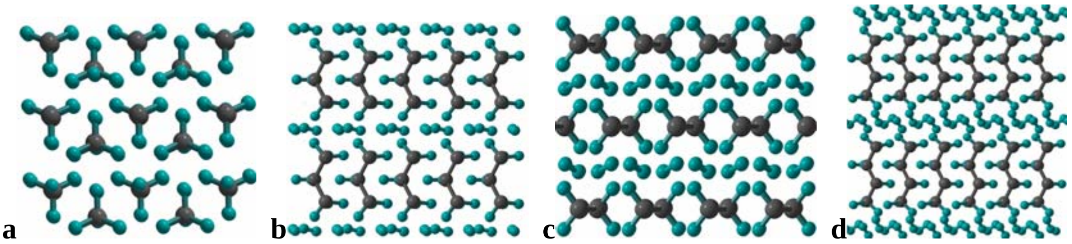


Fig. 12. Structures of CH_4: a) methane structure at 100 GPa, b) C_3H_8*$2H_2$ structure at 200, c) $(CH_2)_\infty$*H_2 structure at 300 GPa, d) C_4H_{10}*$3H_2$ structure at 300 GPa.

Exploration of transition metals at ultrahigh pressures. Many transition metals appear to have the same structure (typically, hcp, fcc or bcc) over a very wide pressure range. This is at first sight surprising, especially when contrasted with the behaviour of simple sp-metals, which often adopt d-character (i.e. become *de facto* transition metals) and a sequence of complex structures on compression. We have performed exploratory simulations of a number of transition metals that at ambient conditions are known in the hcp (Zn, Cd, Ru, Os, Tc, Re), bcc (Mo, W, Nb) and fcc (Cu) structures. For these metals, no phase transitions were experimentally detected up to 126 GPa (Zn), 174 GPa (Cd), 56 GPa (Ru), 75 GPa (Os), 10 GPa (Tc), 251 GPa (Re), 188 GPa (Cu), 272 GPa (Mo), 154 GPa (Nb), 364 GPa (W). Among these metals, only for Os a transition into the hexagonal ω-phase structure (with 6 atoms/cell) was predicted to occur at 840 GPa [67].

The behaviour of each of these metals was explored at several pressures from 0 to 1000 GPa, using system sizes of 2 atoms/cell (Zn), 4 atoms/cell (Zn), 6 atoms/cell (Zn, Cd, Ru, Os, Tc, Re, Nb) and 8 atoms/cell (all metals). We have found that at all pressures up to 1000 GPa (at 0 K), all these metals remain in the same structure as at ambient conditions. This includes Os, for which our calculations find the hcp structure to be much more stable than ω-phase or any other structure at least up to 1000 GPa (Fig. 13). The difference with [67] is likely to be due to pseudopotential errors, but Fig. 13 shows that both PAW and pseudopotential calculations lead to the same conclusion when the pseudopotential is correctly constructed.

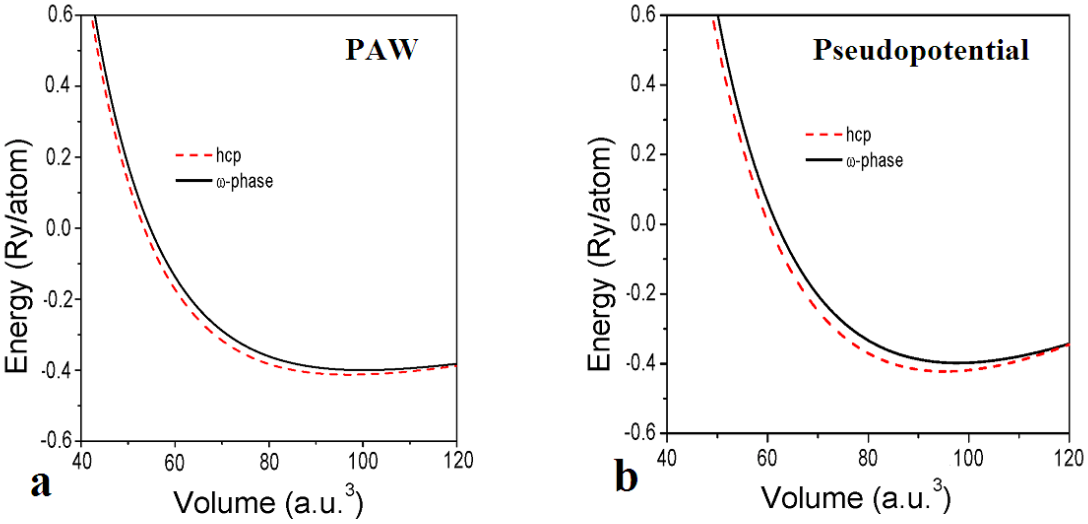


Fig. 13. Energy-volume curves for Os a) PAW results, b) pseudopotential calculations. PAW calculations were performed using the VASP code, pseudopotential results were obtained with the PWSCF code.

Testing the limits of DFT: new hypothetical phases of FeS and FeO. The Fe-S system is important for the understanding of the cores of terrestrial planets (Mercury, Venus, Earth, Mars). Numerous experimental studies of FeS uncovered six solid phases at pressures of up to 100 GPa and temperatures up to 3000 K ([32] and references therein) – all of which are based on the NiAs-type structure with various distortions and superstructures (one of the high-temperature phases has the undistorted hexagonal NiAs structure). Among these phases, two have the same MnP-type structure (*Pnma*-distortion of the NiAs-type structure) – but one is antiferromagnetic (stable between 3.5 GPa and 7 GPa [84]) and the other, recently discovered [32], is non-magnetic and stable above 40 GPa (i.e. at pressures of Martian core). The structure of the non-magnetic MnP-type phase (Fig. 14a) was established in the same study by both Rietveld analysis and evolutionary structure prediction [32].

Tremel et al. [98] have analyzed the nature of various distortions of the NiAs-type structure (assuming non-magnetic ground states), and explained them tracing these distortions in terms of the Jahn-Teller effect and electronic pseudogap formation. These authors found that the

MnP-type structure occurs when the number of valence electrons is 11-14 per formula. FeS, with 14 valence electrons, is therefore at the limit of stability of the MnP structure type and should be prone to phase transitions. The complicated phase diagram of FeS is consistent with this picture; magnetism (not considered in [98]), increases its complexity further. At very high pressures (above 130 GPa, according to GGA calculations) the MnP-type phase was predicted [32] to transform into a new phase. This structure of this seventh phase of FeS (Fig. 14b) belongs to a new structure type, but can be represented as a *Pmmn*-distortion of the NaCl-type structure. FeS is a difficult system for the GGA: while the sequence of phase transitions with pressure is well reproduced by this approximation, the transition pressures are not predicted accurately: most notably, the pressure range where the non-magnetic $P2_1/a$ phase is stable is predicted to be 0-90 GPa (in experiments it is much narrower, 7-40 GPa – see [32] and references therein). Therefore, there is a large uncertainty on the predicted stability range (> 130 GPa) of the *Pmmn* phase of FeS and an experimental determination of this pressure range is desirable, as this phase is potentially important for the cores of the Earth and Venus.

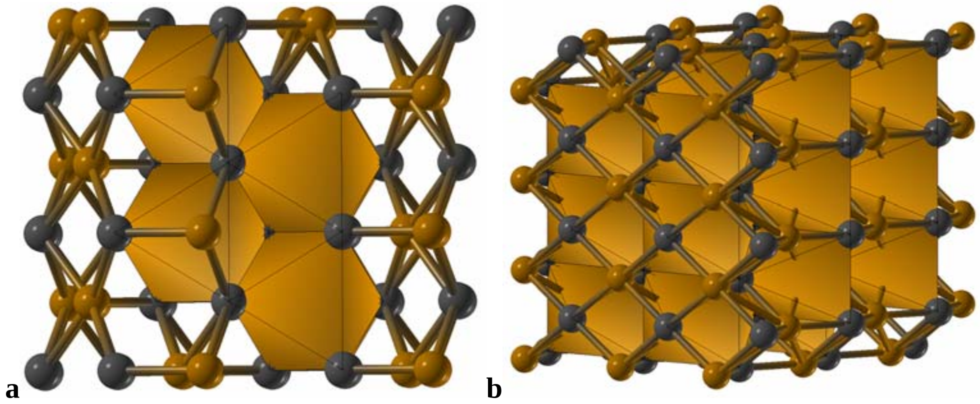


Fig. 14. Structures of high-pressure non-magnetic phases of FeS. a) MnP-type structure (FeS VI), space group *Pnma*; b) distorted NaCl-type structure (FeS VII), space group *Pmmn*. The yellow and black spheres denote iron and sulfur atoms, respectively. FeS6 octahedra are shown.

FeO has long been known to be a system where standard density-functional approximations, the LDA and GGA, fail not only quantitatively but even qualitatively. One of the worst failures is that these approximations predict the wrong ground state at 1 atm – the inverse NiAs-type (i-NiAs) antiferromagnetic phase [85][86] (Fig. 15b). The experimentally observed phase at these conditions has a rhombohedrally distorted antiferromagnetic NaCl-type structure (Fig. 15a), and for this phase LDA and GGA calculations incorrectly predict a metallic behaviour. High-pressure experiments produced strong evidence for the stability of the i-NiAs [88] or NiAs [87][89] structures above ~70-90 GPa.

We performed a series of evolutionary simulations at 100 GPa (with 4,6,8,12 and 16 atoms/cell, all non-magnetic) and found that, within the GGA, the most stable is the unexpected and hitherto unknown trigonal structure (space group $P3_221$, see Fig. 15c). Within the GGA, this structure is stable between 65 GPa and 190 GPa (Fig. 16), and above 190 GPa a reentrant transition into the (non-magnetic) distorted NaCl-type phase is predicted. While these transition pressures are likely to be numerically inaccurate, there is a possibility that the sequence of phases is correct, because at high pressures, where the role of strong on-site Coulomb correlations diminishes and magnetism is suppressed, GGA calculations might produce at least qualitatively correct results (as for FeS). We will not be surprised, however, if the new structure is an artifact of the GGA. Detailed experiments at ultrahigh pressures are needed to address this and assess the accuracy of DFT calculations at ultrahigh pressures.

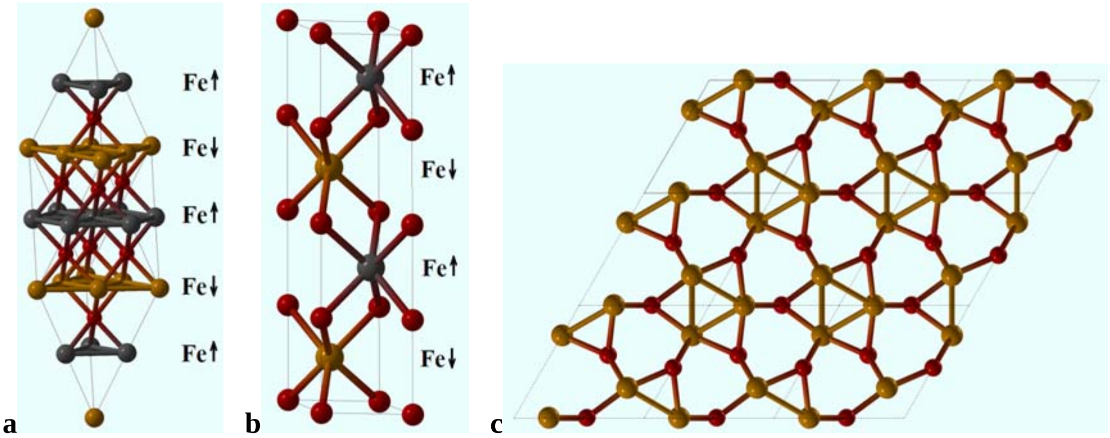


Fig. 15. Phases of FeO. Space group $P3_221$, structure parameters at 100 GPa: a=b=4.5365 Å, c=2.3648 Å, Fe(0; 0.2665; 0.3333), O(0.5564; 0; 0.16667). Distances: Fe-O = 1.77 (x2), 1.80 (x2), Fe-Fe= 2.24 (x2), 2.36 (x2), 2.62 (x2), O-O= 2.36 (x2), 2.44 (x2), 2.64 (x2). In panels a,b Fe atoms with opposite magnetic moments are shown by different colours; layers of atoms with the same magnetic polarization are marked.

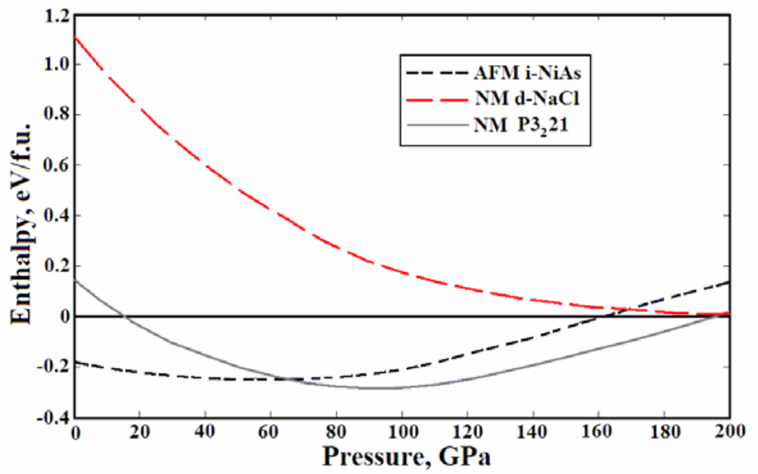


Fig. 16. Stability of FeO phases within the GGA. Enthalpies are relative to the non-magnetic rhombohedrally distorted NaCl-type structure. Spin transition of the NaCl-type is predicted to be gradual and to be completed at ~200 GPa. At pressures between 65 GPa and 190 GPa the new $P3_221$ possesses the lowest theoretical enthalpy. This is possibly a result of shortcomings of the GGA, as well as the prediction of the i-NiAs structure as the ground state at ambient conditions.

Semiconducting and metallic phases of solid oxygen: unusual molecular associations. The red ε-phase of oxygen, stable in the pressure range 8-96 GPa, was discovered in 1979 [55], but its structure was solved only in 2006 [57][58]. The metallic (superconducting at very low temperatures [60]) ζ-phase, stable above 96 GPa, was discovered in 1995 [56], but its structure still remains experimentally unsolved. Neutron diffraction showed [61] that already in the ε-phase (at 8 GPa) there is no long-range magnetic order. The disappearance of magnetism is a consequence of increasing overlap of molecular orbitals with increasing pressure. Ultimately, orbital overlap leads to metallization. To understand high-pressure chemistry of oxygen, we performed extensive structure searches at pressures between 25 GPa and 500 GPa [14][62], restricting ourselves only to non-magnetic solutions.

At 25 GPa we found two particularly interesting structures – one consisting of zigzag chains of O_2 molecules (*Cmcm* structure of Ref.[64][14], see Fig. 17b) and one with more complex chains of molecules (see Fig. 17c). These have strong similarities with the experimentally observed structure (Ref. [57][58], see Fig. 17a) consisting of O_8 clusters: all of these structures are molecular, and in all of them each molecule is connected with two other molecules, at distances of ~2.1-2.2 Å (the intermolecular distance is ~1.2 Å). The *Cmcm* structure, first suggested in [64], is the true GGA ground state, but it differs from experiment; as Fig. 18a shows, its enthalpy is ~10 meV/atom lower than for the experimentally found structure (Fig. 17a). Metastability of the experimentally studied structure cannot yet be ruled out, but, more likely, this discrepancy is due to deficiencies of the GGA. The $(O_2)_4$ clusters are held together by weak intermolecular covalent bonds: each O_2 molecule has two unpaired electrons occupying two molecular π*-orbitals, and sharing these electrons with neighbouring molecules creates two intermolecular bonds per molecule and a non-magnetic ground state [62][66]. It is well known that DFT-GGA does not perform well for stretched covalent bonds, the root of the problem being in the locality of the exchange-correlation hole in DFT-GGA, whereas the true exchange-correlation hole in such cases is highly delocalized. At high pressure, intermolecular distances decrease, intermolecular bonds become more similar to normal covalent bonds and the true exchange-correlation hole becomes more localized. Therefore, we can apply the GGA with greater confidence for the prediction of the structure of the metallic ζ-phase.

For the ζ-phase, evolutionary simulations at 130 GPa and 250 GPa uncovered two interesting structures with *C*2/*m* and *C*2/*c* space groups [62]. These have very similar enthalpies (Fig. 18a); the *C*2/*m* structure is slightly lower in enthalpy and matches experimental X-ray diffraction and Raman spectroscopy data very well, better than the *C*2/*c* structure [62]. Both structures contain well-defined O_2 molecules; our simulations show that oxygen remains a molecular solid at least up to 500 GPa. Phonon dispersion curves of the *C*2/*m* structure (Fig. 18b-d) contain clearly separated molecular vibrons and show that the structure is dynamically stable, except at 110 GPa, where we see tiny imaginary frequencies in the Γ-V direction, close to the Brillouin zone centre – such soft modes may result in small-amplitude long-wavelength modulations of the structure at very low temperatures.

The ε-ξ transition is isosymmetric, which implies that it is first-order at low temperatures but can become fully continuous above some critical temperature [65]. Given the small volume discontinuity upon transition and small hysteresis (one can obtain the *C*2/*m* structure of the ξ-phase by simple overcompression of the ε-O_8 structure, ~5 GPa above the thermodynamic transition pressure), one can expect this critical temperature to be rather low. We note that within the GGA the ε-ξ transition is predicted to occur at 45 GPa, much lower than the experimental transition pressure (96 GPa). This has two explanations – (i) as the GGA is expected to perform better for the metallic ζ-phase than for the semiconducting ε-O_8 phase, the enthalpy differences are expected to suffer from non-canceling errors, (ii) since the ε-ξ transition is not only structural, but also electronic (insulator-metal transition), one might expect metallization at lower pressures than in experiment.

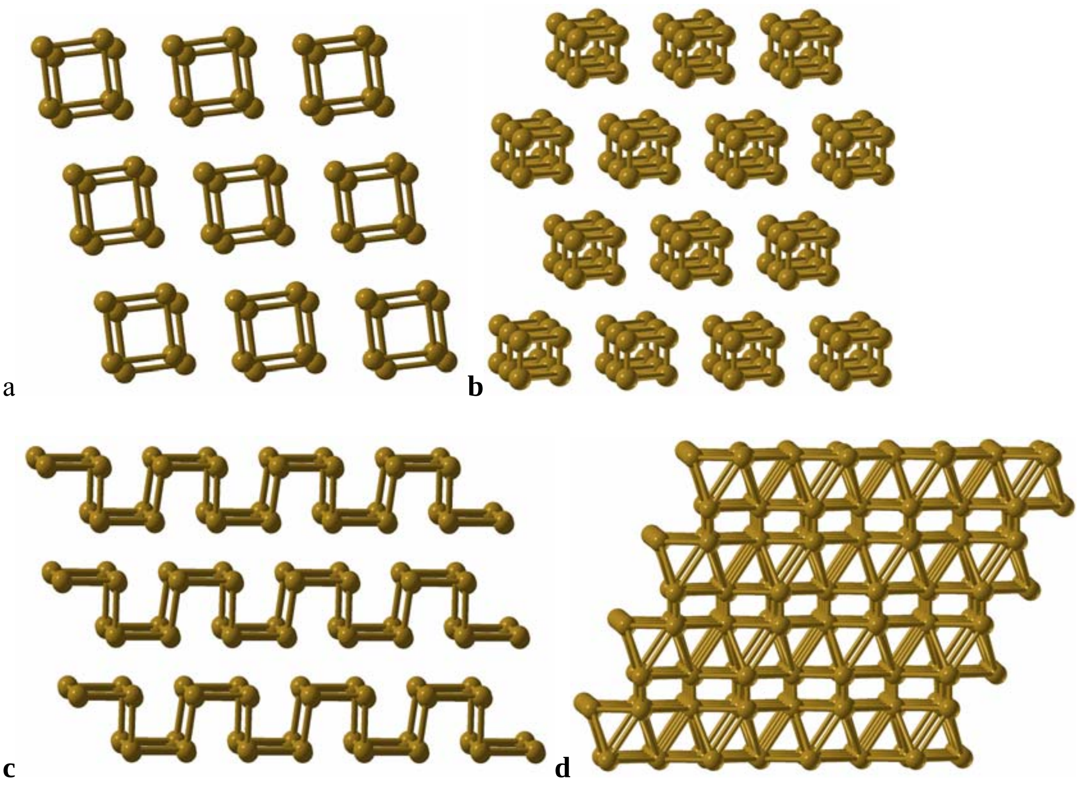


Fig. 17. High-pressure structures of oxygen: a) experimentally found ε-O_8 structure at 17.5 GPa [57], b) *Cmcm* chain structure [64][14], c) metastable chain structure at 25 GPa [14], d) *C*2/*m* structure of the ζ-phase at 130 GPa [62]. Contacts up to to 2.2 Å are shown as bonds.

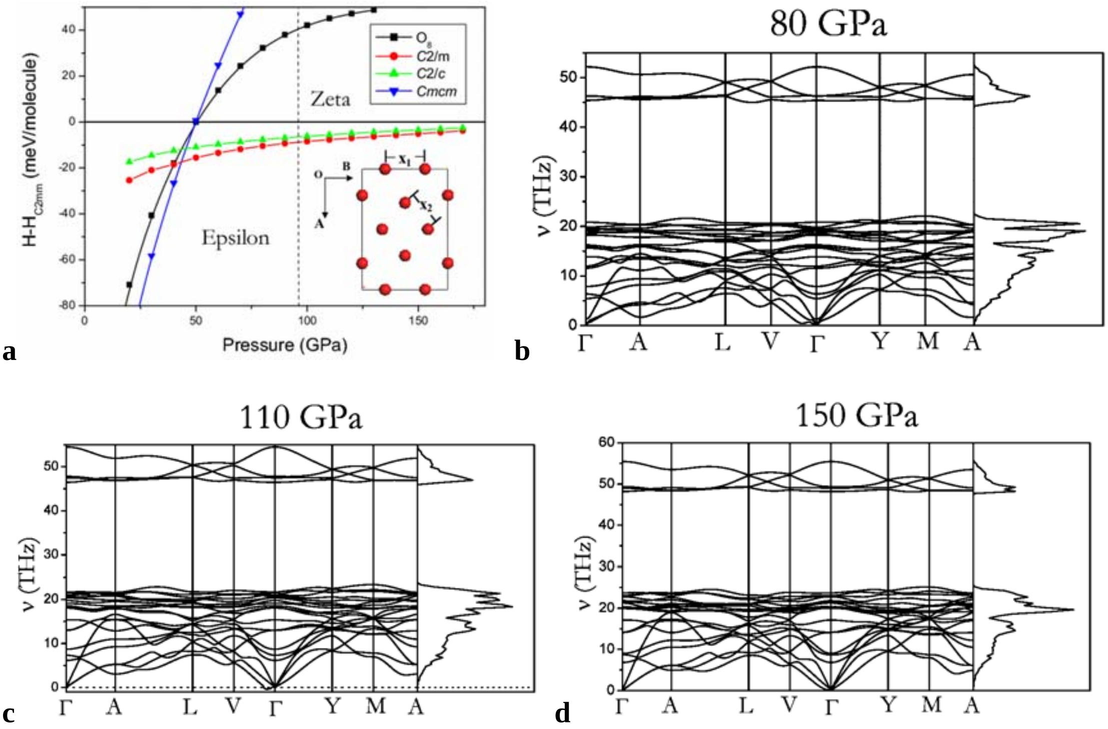


Fig. 18. High-pressure phases of oxygen: a) enthalpies (relative to the *C*2*mm* structure of Ref. [63]) of several possible structures as a function of pressure (from [62]) of several possible structures, b) phonon dispersion curves and densities of states of the *C*2/*m* ζ-phase at three pressures.

Reactivity of noble gases: are Xe-C compounds possible at high pressure? USPEX can be used also for studying multicomponent stoichiometric systems and their reactivity. Grochala [43] has suggested the possibility of stable Xe-C compounds at high pressure; indeed, carbon and xenon have similar valence orbital energies (*cf.* ionisation potentials of 12.13 eV and 11.26 eV for Xe and C, respectively) and one expects that pressure would make Xe more reactive [41]. We did simulations at 200 GPa, i.e. above the metallization pressure of Xe (132 GPa [77]), when its closed electronic shells are strongly perturbed. These calculations were done within the GGA [38] and on cells containing up to 14 atoms/cell. At this pressure all Xe carbides are extremely unstable (Fig. 19), but XeC_2 has a small negative volume of formation and might become stable at much higher pressures. All structures (Fig. 20) are made of close-packed Xe layers (i.e. fragments of the elemental Xe structure) and 3,4-connected carbon layers (intermediate between graphite and diamond), except the 3D-clathrate structure of XeC_8. The observed layering is, most likely, system size-dependent: increasing the number of atoms, we expect to see thicker elemental layers, to the point of complete phase separation. The presence of three-coordinate carbon and a clathrate structure at such high pressure can be explained by an effective negative pressure, which large Xe atoms exert on the much smaller C atoms. Some of the carbon layers are shown in Fig. 21; they are reminiscent of the low-pressure chemistry of carbon. Recently, we succeeded in predicting new stable oxides and silicates of xenon at much lower pressures (Jung *et al.*, in prep.). It seems that high electronegativity of the partner atom is more important for stability of noble gas compounds than similarity of orbital energies.

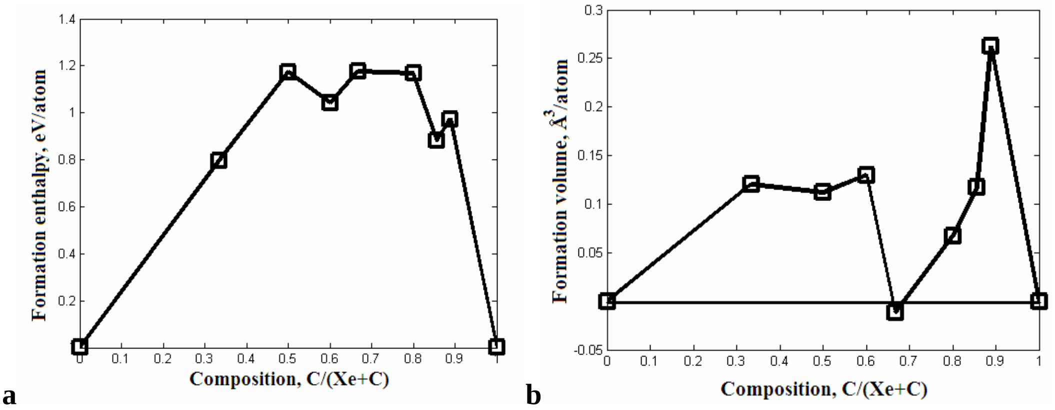


Fig. 19. Predicted enthalpy (a) and volume (b) of formation of Xe-C compounds at 200 GPa. The compounds shown are Xe (hcp), Xe_2C, XeC, Xe_2C_3, XeC_2, XeC_4, XeC_6, XeC_8, C(diamond).

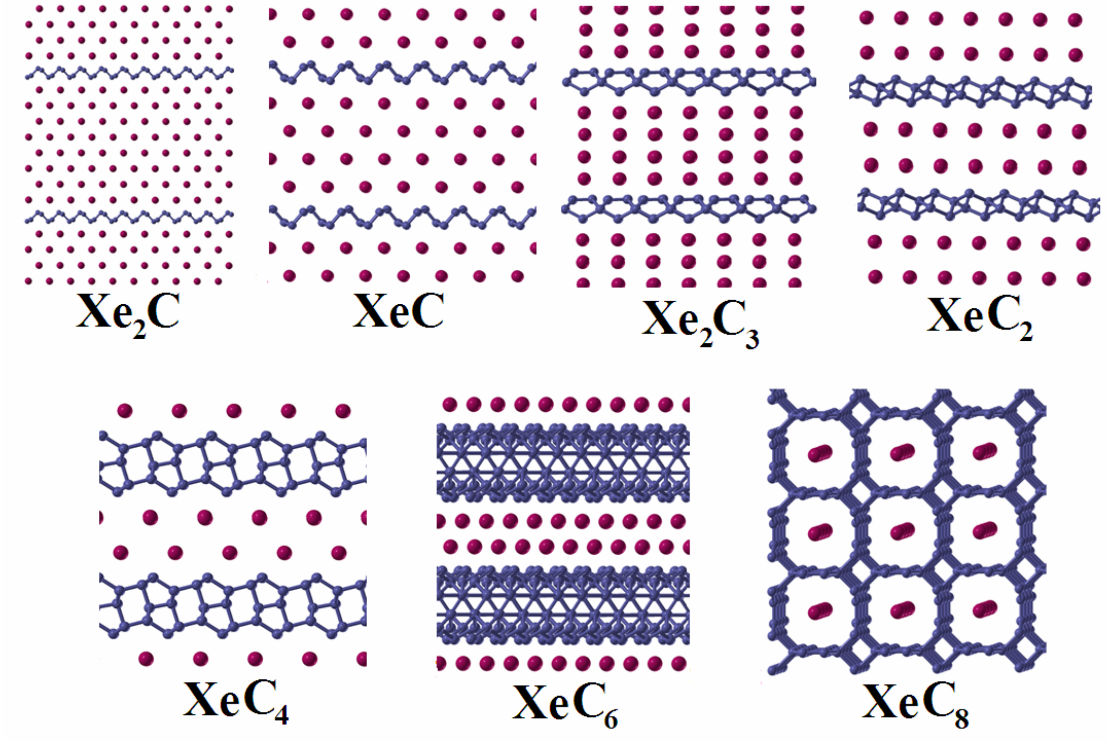


Fig. 20. Predicted structures of Xe_2C, XeC, Xe_2C_3, XeC_2, XeC_4, XeC_6, XeC_8 at 200 GPa.

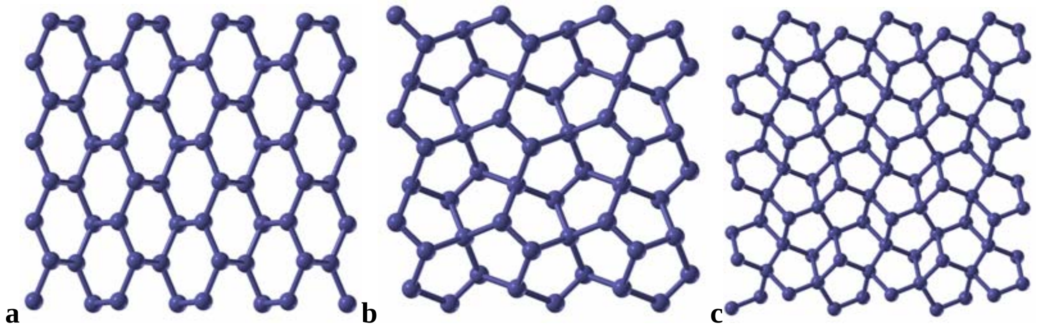


Fig. 21. Structures of carbon layers in a) Xe_2C and XeC, b) Xe_2C_3, c) XeC_2.

Another example of unmixing: Cu-C system. While many metals easily form carbides already at ambient pressure, no stable compounds are known in the Cu-C system. The tendency to unmixing in this system is very strong and even simulations on small cells show clear separation into layers of fcc-structured Cu and layers of graphite (Fig. 22). When the tendency to unmixing is not so large, simulations on small unit cells may find metastable "mixed" structures (see below, Fig. 23c). Such structures have the lowest thermodynamic potential only at the given number of atoms in the unit cell; increasing the cell size would lead to phase separation. In the Cu-C system, phase separation is evident already at very small system sizes (Fig. 22).

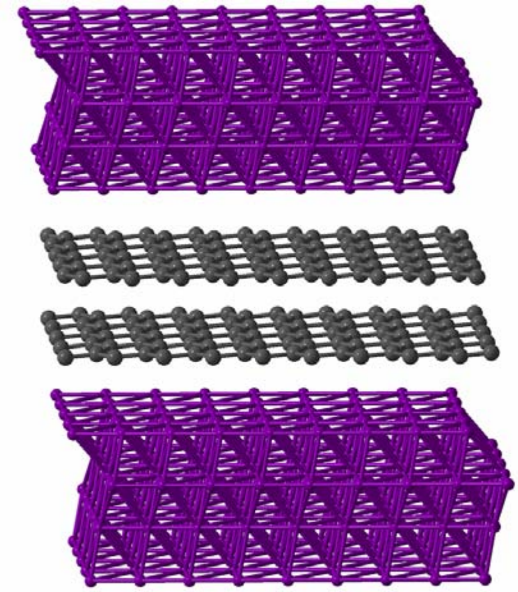


Fig. 22. Lowest-energy structure of Cu_2C with 12 atoms/cell at 1 atm.

Investigation of possible insulating phases of $Al_{12}C$ and $Al_{13}K$. Works [35]-[37] proposed a new class of solids, made of Al clusters – ionic insulator $(Al_{13})K$ with the CsCl-type structure and icosahedral [35] or cuboctahedral [36] Al_{13}^- ions, and molecular $Al_{12}C$ with icosahedral [37] $Al_{12}C$ clusters bound together by weak intermolecular interactions. Icosahedral nanoclusters are well known for aluminium, boron, and other elements, and according to the jellium model, clusters with 40 valence electrons should be particularly stable and $Al_{12}C$ and Al_{13}^- icosahedra groups may have closed-shell electronic configurations and produce insulating structures. Such materials, if they can be synthesized, would be technologically very interesting materials. Recently [69] we explored these systems (with the exception that for $Al_{12}C$ we study only the 13-atom system, rather than the previously [37] suggested 104-atom cell) using USPEX. In both cases the lowest-energy structures do not contain any well-defined clusters.

For $Al_{13}K$, when full relaxation is performed, the icosahedral structure (Fig. 23a) spontaneously transforms into the cuboctahedral one (Fig. 23b). The cuboctahedra are fragments of the fcc structure; however, these are not isolated in $Al_{13}K$, but form bonds with each other (Al-Al distances within the cluster are 2.78 Å, and 2.72 Å between the clusters), which makes this structure metallic and indicates limitations of the jellium model. The structure found with USPEX (Fig. 23c) is 78 meV/atom lower in energy, has no clusters and shows a strong perturbation exerted by K atoms on the Al-sublattice. No stable compounds are known in the Al-K system – this is consistent with our result that even the USPEX-produced structure is 72 meV/atom higher in energy than the mechanical mixture of pure Al and K.

For $Al_{12}C$, the optimized icosahedral structure (Fig. 24a) also has bonds between the clusters: Al-Al distances are 2.67-2.72 Å within the cluster and 2.65 Å between the clusters. Thus, the expectation of a molecular structure made on the basis of the jellium model is again incorrect. This structure has a DFT band gap of 0.3 eV, but is not stable: with USPEX, we found a much more stable (by 557 meV/atom) structure shown in Fig. 24b. That structure is close-packed, similar to the fcc structure of pure Al, but with stacking faults at which C atoms occupy the octahedral voids in the aluminium close packing. Thus, C impurities in Al may be correlated with the presence of stacking faults. The USPEX-produced structure of $Al_{12}C$ is metallic and only 39 meV/atom less stable than the mechanical mixture of pure aluminium and graphite. There is a stable phase in the Al-C system, Al_4C_3.

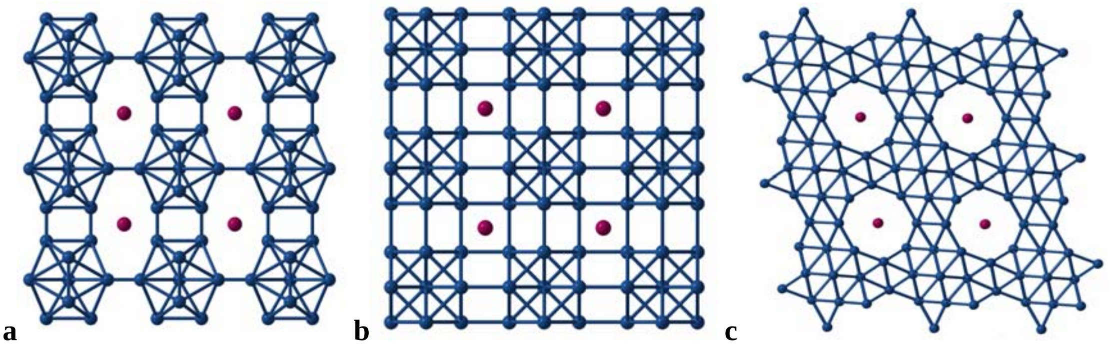


Fig. 23. Structures of $Al_{13}K$ with 14 atoms/cell: (a) with icosahedral Al_{13} clusters, (b) with cuboctahedral Al_{13} clusters, (c) structure produced by USPEX. From [69].

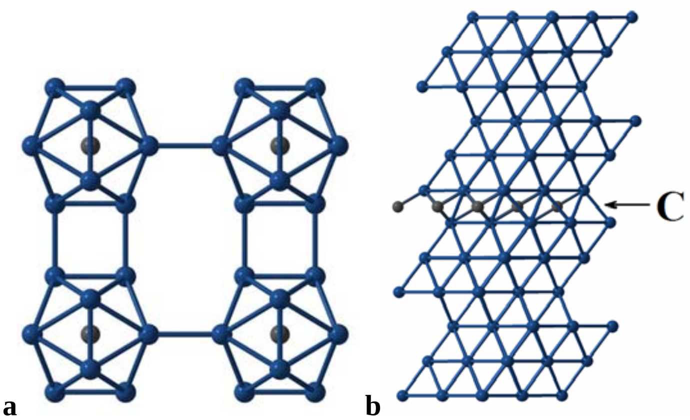


Fig. 24. Structures of $Al_{12}C$ with 13 atoms/cell: (a) with icosahedral clusters, (b) structure produces by USPEX. From [69].

5 Conclusions

Evolutionary algorithms, based on physically motivated forms of variation operators and local optimization, are a powerful tool enabling reliable and efficient prediction of stable crystal structures. This method has a wide field of applications in computational materials design (where experiments are time-consuming and expensive) and in studies of matter at extreme conditions (where experiments are very difficult or sometimes impossible). We are suggesting USPEX as the method of choice for crystal structure prediction of systems with ~ 6 – 40 atoms/cell, where no information (or just the lattice parameters) is available. Above 40 atoms/cell runs become expensive (although still feasible), eventually necessitating the use of other ideas within USPEX or another approach, due to the 'curse of dimensionality'. Fortunately, many (or most) systems of practical or fundamental interest fall in the size range accessible to USPEX. The extension of the method to molecular systems (i.e. handling whole molecules, rather than individual atoms) is already available. One of the current limitations is the accuracy of today's *ab initio* simulations; this is particularly critical for strongly correlated and for van der Waals systems. Note, however, that the method itself does not make any assumptions about the way energies are calculated and can be used in conjunction with any method that is able to provide total energies. Difficult cases are aperiodic and disordered systems (for which only the lowest-energy periodic approximants and ordered structures can be predicted at this moment). Additional work is required to extend it further to very large (> 200 atoms/cell) systems and to systems with variable composition. In the latter case, one would simultaneously predict all the stable stoichiometries in a given range of compositions, and the corresponding stable structures. A pioneering study [90] succeeded in predicting stable stoichiometries of alloys within a given structure type, but simultaneous prediction of structure and stoichiometry remains an unsolved problem. Currently, we are working on such extensions of the method.

Acknowledgements

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Appendix: How to compute the similarity matrix. Fingerprinting method.

Here we describe a method which one of us (M.V.) has developed to monitor population diversity in evolutionary simulations. To measure the similarity between the structures, we propose a clustering procedure based on some definition of the abstract distance between two structures. Any such method should be: 1) independent from translation and rotation of the structure; 2) independent from the choice of unit cell between equivalent unit cells; 3) robust regarding small numerical errors; 4) independent from the ordering of cell axis and atoms in the cell; 5) independent from inversions and mirroring of the structures.

Among the number of papers (e.g. [91]-[94]) where structure descriptors and distance metrics were considered for crystals, we note the work of Chisholm and Motherwell [94], which employed interatomic distances and provided inspiration for our more general method. The work of Hundt et. al. [93] gives a comprehensive survey of existing methods with a focus on calculating some form of distance metric between structures.

Using this distance, we construct a similarity matrix where each element A_{ij} measures the similarity (in terms of the abstract distance) between the i-th and j-th structures. We prefer to rescale the abstract distances, so that for a maximally similar pair the similarity equals one, and for the most dissimilar pair it is zero.

Properties of similarity matrices:

1. The range of values is from 0 (blue) to 1 (red).
2. Similarity matrix is symmetric, with values of 1 on the main diagonal.
3. Blue lines indicate structures that have no significant similarity with any other structures explored.
4. Blocks of values close to the main diagonal show diversity of the population and its change along the simulation. Due to selection pressure, in evolutionary simulations diversity of the population decreases during the simulation – visually this can be detected as "reddening" of the similarity matrix towards its upper right corner.

To calculate the distance between the structures, a descriptor, called a fingerprint, is computed for each structure. The fingerprint can be defined in the real space (set of interatomic distances) or in the reciprocal space (structure factor), we prefer the more intuitive real-space constructions. One (out of an infinite number of possibilities) construction we use consists of components from all element types; each component contains the element type of the corresponding atom and an increasing ordered set of distances from it to the other atoms in an infinite crystal (Fig. 25). All components contain the same number of distances and it is sufficient to have distances up to one half of the longest unit cell side in the examined set of structures.

To cluster fingerprints, and therefore to cluster the associated structures, a distance between them should be defined. We start defining the distance between two fingerprint components. The simplest measure is the Euclidean distance between the N-dimensional points whose coordinates are the various component distances (*da* for first fingerprint and *db* for the second one).

$$D = \sqrt{\sum_i (da_i - db_i)^2}$$

To increase the discriminating power, one should account for the linear increase of errors with distance and the quadratic increase with distance of the number of distance values in the fingerprint, e.g.:

$$D = \sqrt{} \sum_i ((da_i - db_i) / (da_i + db_i)^3)^2$$

This algorithm has been implemented inside the molecular visualization environment STM3 [95][96] and uses its visualization facilities to help guide and control the algorithm behavior. The method uses a library for approximate nearest neighbor searching [97] to compute the number of distances needed for each structure atoms.

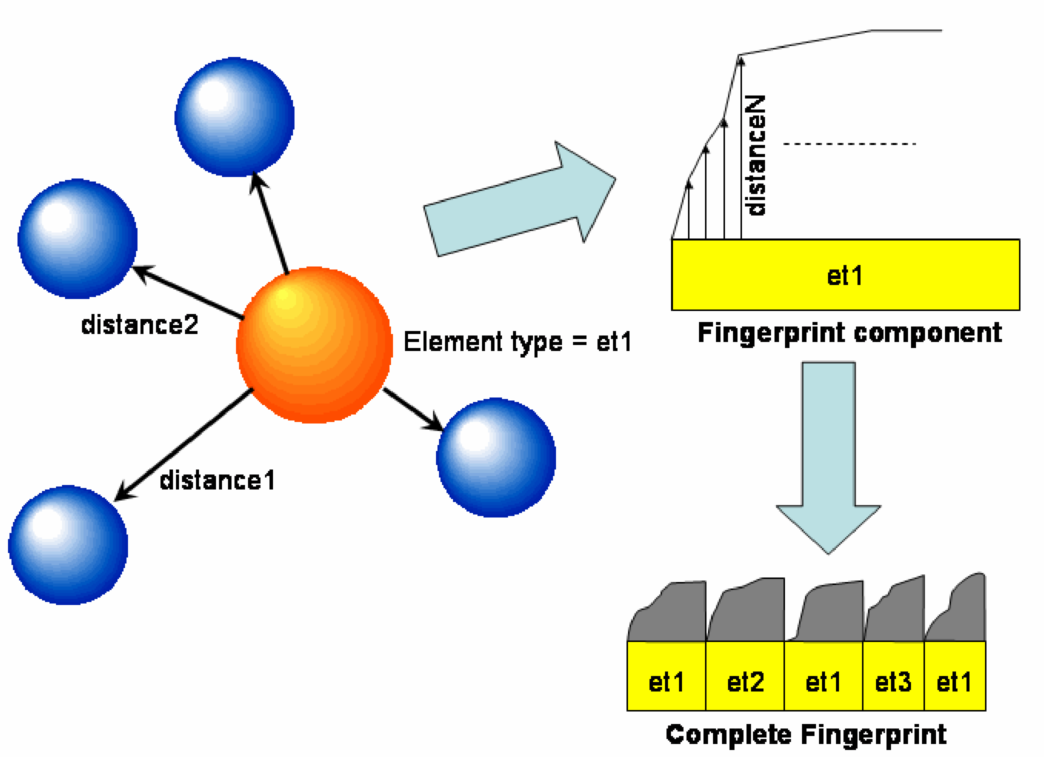

Fig. 25. Structure of a fingerprint.

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