Report on the "Catalysis from First Principles" workshop at CECAM, France
September 11-14 2006

Organisers

Angelos Michaelides (Fritz-Haber-Institut der Max-Planck-Gesellschaft, Germany)
Matthias Scheffler (Fritz-Haber-Institut der Max-Planck-Gesellschaft, Germany)
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I. Summary

More than fifty people were present in Lyon for the fifth "Catalysis from first Principles" meeting which took place from the 11th to the 14th of September.

The workshop highlighted several key applications of first principles electronic structure techniques as well as the combination of electronic structure techniques and statistical mechanics to interesting systems in heterogeneous catalysis and electrochemistry.

A good mix of participants from academia and industry attended the meeting. And also a large fraction of experimentalists attended (~1/3).

The meeting served its purpose well by bringing together experimentalists and theoreticians from industry and academia, as well as having a nice balance of experienced and junior researchers. Areas of common interest were identified between many participants from which future collaborations are likely to follow.

II. Description of the scientific content of and discussion at the event

The properties of surfaces and the processes that occur on them are relevant to many aspects of daily life and scientific endeavour. The efficiency of numerous heterogeneous catalysts and electrochemical devices depends directly, for example, on reactions that occur at precious metal surfaces. The key to future advances in these areas is to understand surface processes at the atomic level. One way to reach this understanding is through first-principles simulations and indeed such simulations have now reached a state of maturity such that they can contribute toward the development of new catalysts or make faithful predictions about any number of surface properties.

Increasingly such first-principles electronic structure approaches are being combined with techniques from statistical mechanics and thermodynamics in order to understand the statistical mechanical interplay of the (typically) many elementary reaction steps that take place “in concert” on the surface of a working (heterogeneous) catalyst. The power of this combined, multiscale modeling, approach is also increasingly being exploited to make predictions about the properties of catalytic substrates at the high pressures and temperatures at which the majority of catalytic processes operate. Successfully combining these techniques and in so doing “bridging” the often-discussed pressure and materials gaps that separate the atomic zero-temperature zero-pressure regime of electronic structure calculations from the more realistic conditions of industrial catalysis remains one of the grand challenges of catalysis research.

One of the aims of the workshop was to bring together leading experts and practitioners from the electronic structure theory and statistical mechanics communities to discuss the prospects and potential for further coupling of these approaches leading to future “first principles catalytic design”. Further, since there is an often symbiotic relationship between experiment and theory in this area, with many sophisticated experimental surface probes relying on theoretical modelling for their quantitative interpretation, experimental surface science and catalysis practitioners were also invited.

Through the more than 20 invited talks from leading paractitioners in each of the above fields the current status of and future prospects for applying ab initio and statistical mechanics approaches to the study of chemical processes at solid surfaces was reviewed and discussed; examples of how calculational methods are now being used as a tool to aid the design of new heterogeneous and electrochemical catalysts were covered; and recent applications and future prospects of using such approaches to understand interfaces of electrochemical and environmental (atmospheric) importance were discussed.

Talks

Most of the workshop involved invited talks of 30 minutes duration, followed by discussion of 15 minutes. The question periods were always lively and constructive, allowing for very fruitful exchange of views. A list of the 22 invited speakers along with the titles of their talks is given below.

W. Henry Weinberg:	Nonconventional Catalysts for Isotactic Propene Polymerization in Solution Developed by Using High-Throughput-Screening Technologies
Thomas Bligaard:	Catalysis Informatics and the methanation reaction
Florian Mittendorfer:	The morphology of mesoscopic metal clusters under oxidizing conditions
Jutta Rogal:	A first-principles statistical mechanics investigation of the stability of a thin surface oxide in reactive environments
William F. Schneider:	Environmental Catalysis at the Boundary between Metals and Metal Oxides

Michel Daage:	Modeling Catalysts: An Experimentalist point of view
Peijun Hu:	O_2 activation on Au/TiO_2 and NO dissociation on Ag/Al_2O_3: A DFT study on the interface chemistry
Joachim Sauer:	Treating Dispersion Effects in Extended Systems by Hybrid MP2:DFT calculations: Protonation of Hydrocarbons in Zeolites
Axel Gross:	Catalytic properties of bimetallic substrates and electrodes
Dieter M. Kolb:	Identifying reaction sites in electrocatalysis
Giancarlo Cicero:	Structural properties of water confined between hydrophilic and hydrophobic surfaces, as probed by ab-initio molecular dynamics
Jan Rossmeisl:	Activity of Fuel Cell Electrode Materials
Jens Rostrup-Nielsen:	Step Sites in Syngas Catalysis. The Multiple Approach
Charles T. Campbell:	Thermodynamics of Surface Reactions: Relationship to Reactivity
Jens Nørskov	
Olaf Deutschmann:	Coupling of chemical reactions with mass and heat transport in heterogeneous catalysis
Jesper N. Andersen:	Shining light on surface structures and reactions

André Fielicke: Probing the structures of pure metal clusters and metal cluster complexes

Stephen Jenkins: Towards Chiral Catalysis at Surfaces: Structure, Symmetry and Stereograms

Joachim Paier: Hybrid functionals applied to extended systems

Philippe Sautet: Catalytic active sites and molecular reactivity from a DFT approach

Thomas Risse: On the influence of point defects on the properties of deposited metal atoms and clusters on oxides

Posters

A poster session was held on the second evening of the meeting at which 24 contributed posters were presented, the titles and authors of which are given below.

Photoelectrochemical Hydrogen Production - Selective transition metal substitution of Anatase Titania
by Nicholas Lambropoulos

Theoretical analysis of XAS spectra of Ti compounds as catalysts in hydrogen absorption/desorption reactions
by Takao Tsumuraya, Tatsuya Shishidou,Tamio Oguchi

Adsorption and photocatalytic activity of perylene diimide based light harvesting molecules over Pt surfaces
by Oguz Gulseren, E. Mete, S. Ellialtioglu, D. Uner

Electronic structure of visible light-active N-doped TiO2 photocatalyst
by José C. Conesa

Dissociation of NO and recombination of oxygen atoms on Ag(100)
by Mahesh Menon, R. J. Buenker, J. L. Whitten

First principles study of the dehydrogenation NHx and the reverse reactions on Rh(111) surface
by Cristina Popa, R. A. van Santen and A. P. J. Jansen

Ammonia decomposition in N-doped carbon nanotubes
by Antonio José Roque da Silva, Mariana Rossi, A. Fazzio

Investigation of the reaction between Aluminum cluster and Methane
by Helen Alexandrou, N.C. Bacalis

Ab-initio calculations of free-energy reaction barriers: Application to monomolecular cracking of hydrocarbons over zeolites
by Tomas Bucko, Ch. Dellago, J. Hafner, L.Benco, and J. Angyan

Epoxidation of Phenylpropenes on Cu(111): DFT Cluster Studies on Geometry and C 1s NEXAFS Spectra of the Adsorbate Species
by Klaus Hermann, C. Kolczewski, M. Cavalleri, F. J. Williams, R. L. Cropley, and R. M. Lambert

Theoretical calculations of partially reduced ceria surfaces
by Kersti Hermansson, B. Herschend, M. Baudin and Z. Yang

Extended Ab-Initio Atomistic Thermodynamics for Electrochemical Systems
by Timo Jacob, Matthias Scheffler

Promoter segregation in Co(Ni)MoS catalysts: a DFT study
by Emmanuel Krebs, Pascal Raybaud, B. Silvi

"Textbook" adsorption on "nontextbook" adsorption sites: Halogen atoms on alkali halides surfaces
by Bo Li, A. Michaelides, and M. Scheffler

Computer-assisted search for new ammonia synthesis catalysts
by Ture Munter, Thomas Bligaard, Jeffrey Greeley, Claus H. Christensen, and Jens K. Nørskov

Chemical environment and size effects on Pt catalyst properties: an ab-initio study
by Byungchan Han, C. R. Miranda and G. Ceder

Role of surface topology of MgO on the properties of OH groups: a combined theoretical and experimental study
by Céline Chizallet, Guylène Costentin, Hélène Lauron-Pernot, Jocelyne Maquet, Françoise Delbecq, Philippe Sautet, Michel Che

Theoretical Insight of Adsorption Thermodynamics and Soft Vibrations of Multifunctional Molecules on Metal Surfaces
by David Loffreda

Influence of the hydroxylation and metal coverage of ã-Al2O3 surfaces on the nucleation and growth of Pdn clusters (n=1,5): a DFT study
by Pascal Raybaud, Manuel CORRAL VALERO and Philipe Sautet

Single Molecule's Inelastic Spectroscopy on metal surfaces: Insight from DFT
by Marie-Laure Bocquet, Hervé Lesnard and Nicolas Lorente

Compensation Effect in Kinetic Monte-Carlo Simulations of Heterogeneous Catalysis
by Hakim Meskine, Karsten Reuter and Matthias Scheffler

Competitive routes for a chemo-regioselective reaction on a metal surface from a theoretical approach.
by Françoise Delbecq, D. Loffreda, F. Vigné, P. Sautet

CP2K/Quickstep: Accurate Periodic Calculations Using Large Gaussian Basis Sets
by Matthias Krack

When seeing is not believing: Oxygen on Ag(111), a simple adsorption system?
by A. Michaelides, K. Reuter, and M. Scheffler

III. Assessment of the results and impact of the event on the future direction of the field

The field is expanding and lot's of exciting science is being performed! This workshop series should continue. A follow-up meeting in Wien in 2008 was discussed.

Yet closer contacts between experimentalists and theoreticans in this field are to be encouraged.

Yet closer contacts between industrialists and academics in this field are to be encouraged.

IV. Final Programme

Monday, September 11

14:00 – 14:50	Registration	
14:50 – 15:00	**Matthias Scheffler**	Welcome
Chair: Matthias Scheffler		
15:00 – 15:45	**W. Henry Weinberg**	Nonconventional Catalysts for Isotactic Propene Polymerization in Solution Developed by Using High-Throughput-Screening Technologies
15:45 – 16:30	**Thomas Bligaard**	Catalysis Informatics and the methanation reaction
16:30 – 17:00	Coffee & Discussion	
17:00 – 17:45	**Florian Mittendorfer**	The morphology of mesoscopic metal clusters under oxidizing conditions
17:45 – 18:30	**Jutta Rogal**	A first-principles statistical mechanics investigation of the stability of a thin surface oxide in reactive environments

Tuesday, September 12

Chair: Karsten Reuter		
9:30 – 10:15	**William F. Schneider**	Environmental Catalysis at the Boundary between Metals and Metal Oxides
10:15 – 11:00	**Michel Daage**	Modeling Catalysts: An Experimentalist point of view
11:00 – 11:30	Coffee & Discussion	
11:30 – 12:15	**Peijun Hu**	O_2 activation on Au/TiO_2 and NO dissociation on Ag/Al_2O_3: A DFT study on the interface chemistry
12:15- 13:00	**Joachim Sauer**	Treating Dispersion Effects in Extended Systems by Hybrid MP2:DFT calculations: Protonation of Hydrocarbons in Zeolites
13:00 – 14:30	Lunch	
Chair: Jens Nørskov		
14:30 – 15:15	**Axel Gross**	Catalytic properties of bimetallic substrates and electrodes
15:15 – 16:00	**Dieter M. Kolb**	Identifying reaction sites in electrocatalysis
16:00 – 16:30	Coffee & Discussion	
16:30 – 17:15	**Giancarlo Cicero**	Structural properties of water confined between hydrophilic and hydrophobic surfaces, as probed by *ab-initio* molecular dynamics
17:15 – 18:00	**Jan Rossmeisl**	Activity of Fuel Cell Electrode Materials
18:00 –	**Poster Session & Buffet**	

Wednesday, September 13

Chair: Thomas Bligaard		
9:30 – 10:15	**Jens Rostrup-Nielsen**	Step Sites in Syngas Catalysis. The Multiple Approach
10:15 – 11:00	**Charles T. Campbell**	Thermodynamics of Surface Reactions: Relationship to Reactivity
11:00 – 11:30	Coffee & Discussion	
11:30 – 12:15	**Jens Nørskov**	
12:15- 13:00	**Olaf Deutschmann**	Coupling of chemical reactions with mass and heat transport in heterogeneous catalysis
13:00 – 14:30	Lunch	
Chair: Angelos Michaelides		
14:30 – 15:15	**Jesper N. Andersen**	Shining light on surface structures and reactions
15:15 – 16:00	**André Fielicke**	Probing the structures of pure metal clusters and metal cluster complexes
16:00 – 16:30	Coffee & Discussion	
16:30 – 17:15	**Stephen Jenkins**	Towards Chiral Catalysis at Surfaces: Structure, Symmetry and Stereograms
17:15 – 18:00	**Joachim Paier**	Hybrid functionals applied to extended systems
19:30…	**Conference dinner at Brasserie "le Nord"**	

Thursday, September 14

Chair: Charles T. Campbell		
9:30 – 10:15	**Philippe Sautet**	Catalytic active sites and molecular reactivity from a DFT approach
10:15 – 11:00	**Thomas Risse**	On the influence of point defects on the properties of deposited metal atoms and clusters on oxides
11:30 – 12:00	Discussion, close and coffee	

V. Participant List

Australia	Nicholas Lambropoulos (CSIRO, Australia)
Austria	Florian Mittendorfer (Universität Wien)
	Joachim Paier (Universität Wien)
	Tomas Bucko (Universität Wien)
Brazil	Antonio José Roque da Silva (Universidade de São Paulo)
Denmark	Thomas Bligaard (Technical University of Denmark)
	Jens Rostrup-Nielsen (EURAB, Haldor Topsoe)
	Jan Rossmeisl (Technical University of Denmark)
	Ture Munter (Technical University of Denmark)
France	Philippe Sautet (Ecole Normale Supérieure de Lyon)
	Nicolas Dinter (IFP)
	David Loffreda (Ecole Normale Supérieure de Lyon)
	Françoise Delbecq (Ecole Normale Supérieure de Lyon)
	Marie-Laure Bocquet (Ecole Normale Supérieure de Lyon)
	Pascal Raybaud (IFP)
	Emmanuel Krebs (IFP)
	Céline Chizallet (Université Pierre et Marie Curie)
Germany	Axel Gross (University of Ulm)
	Jutta Rogal (Fritz-Haber-Institut der Max-Planck-Gesellschaft)
	André Fielicke (Fritz-Haber-Institut der Max-Planck-Gesellschaft)
	Dieter M. Kolb (University of Ulm)
	Thomas Risse (Fritz-Haber-Institut der Max-Planck-Gesellschaft)
	Joachim Sauer (Humboldt University, Berlin)
	Olaf Deutschmann (Universität Karlsruhe)

	Hakim Meskine (Fritz-Haber-Institut der Max-Planck Gesellschaft)
	Timo Jacob (Fritz-Haber-Institut der Max-Planck-Gesellschaft)
	Bo Li (Fritz-Haber-Institut der Max-Planck-Gesellschaft)
	Klaus Hermann (Fritz-Haber-Institut der Max-Planck-Gesellschaft)
	Matteo Cavalleri (Fritz-Haber-Institut der Max-Planck Gesellschaft)
Greece	Helen Alexandrou (National Hellenic Research)
Israel	Mahesh Menon (Weizmann Institute of Science)
Italy	Giancarlo Cicero (Polytechnic of Torino)
Japan	Takao Tsumuraya (Hiroshima University)
Netherlands	Cristina Popa (Eindhoven University of Technology)
Spain	José C. Conesa (Instituto de Catálisis y Petroleoquímica, CSIC)
Sweden	Jesper Andersen (Lund University)
	Kersti Hermansson (Uppsala University)
Switzerland	Matthias Krack (ETH Zurich)
Turkey	Oguz Gulseren (Bilkent University)
United Kingdom	Peijun Hu (The Queen's University of Belfast)
	Stephen Jenkins (University of Cambridge)
	Sam French (Johnson Matthey TC)
United States	William F. Schneider (University of Notre Dame)

Charles T. Campbell (University of Washington)
W. Henry Weinberg (Symyx Technologies, Inc.)
Michel Daage (ExxonMobil)
Byungchan Han (Massachusetts Institute of Technology)
J. Douglas Kushnerick (ExxonMobil)