Research Networking Programmes

Science Meeting – Scientific Report

***Proposal Title**: Basic Concepts and First-Principles Computations for Surface Science: Applications in Chemical Energy Conversion and Storage*

Application Reference N°: 4450

1) Summary

The objective of this workshop was to discuss recent advances in electronic structure theory in the diverse application areas of surface science. The idea was to provide an overview over state-of-the-art concepts and a perspective on current frontiers and future developments. For this purpose world-leading experts in the different fields delivered a total of 20 extensive 60-min tutorial lectures, as well as eleven 30-min hot topic talks that highlighted current frontier research and open issues. A copy of the final program is part of this report. Conceptually, the program centred on four pillars that together form the scaffold for the present-day operation of first-principles scientists in surface catalysis and basic energy research:

- **Fundamentals and challenges of surface electronic structure calculations**
- **Techniques for the 1st-principles calculation of key quantities**
- **Integration of 1st-principles data into statistical mechanics**
- **1st-principles modelling for the quantitative interpretation of experiments**

In total, 71 participants from more than 15 countries attended the workshop. To further encourage the active participation of younger scientists two extended poster sessions with a total of 41 poster presentations were held on two evenings. With the quaint setting on Norderney, a program structure with ample time for discussions, and a conference outing attended by all participants the workshop aimed for extensive interaction between senior and junior scientists, as well as networking possibilities. The unanimous feedback in particular from the younger participating scientists confirms that this aim has been fully accomplished.

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2) Description of the scientific content of and discussions at the event

Electronic and atomistic processes of molecules at solid surfaces play a central role for a wealth of present and emerging technology. The concomitant conversion of materials, molecules, or energy critically determines functionalities of sensors, catalysts, organic solar or galvanic cells. First-principles calculations have become a key contributor to understanding and new developments in these various application areas. Yet, not least due to this success, the richness and complexity of corresponding calculations has grown at a staggering pace, and so has the development of specific concepts in the different fields.

Due to this interdisciplinarity, also the background and scientific communities of the lecturers and participants were quite diverse. It was therefore a core objective of the workshop (and the program structure based on extensive tutorial-type lectures) to familiarize the participants with the different subjects, to encourage interdisciplinary interactions, and to share experience of different research fields with one another. In this way, we managed to foster the exchange of ideas and methods, to highlight the apparent and the hidden similarities of different systems and approaches, and hopefully stimulated new and fruitful collaborations across subject boundaries.

The program started with introductions into the frontiers of electronic-structure theory calculations, and their specific implementations for surface calculations, given by Matthias Scheffler and Luca Ghiringhelli (FHI Berlin). A key concern and current bottleneck is the achievable accuracy at tractable numerical costs, as well as the modeling of the active sites structure at the surface. For the characterization of the latter, calculation efforts need to go hand in hand with experimental surface science work, an overview of which from the experimental perspective was presented by Martin Weinelt (FU Berlin) – and on the consecutive evening from the view of computational spectroscopy by Patrick Rinke (FHI Berlin). The other Monday sessions focused on Molecular Nanoscience and Materials for Energy, discussing as important concepts as self-assembly (Christof Wöll, KIT Karlsruhe), the treatment of dispersive interactions (Alex Tkatchenko, FHI Berlin), and computational screening (Nicola Marzari, EPF Lausanne). The talk by Alexandra Vojvodic (Stanford, USA) covered corresponding work (and scaling relation concepts) in the catalysis context, thereby bridging to the talks in the Energy to Kinetics session on Tuesday morning. In these talks, the focus shifted away from the actual electronic structure calculations themselves to the way how corresponding information enters into explicit kinetic modeling. Specifically, Graeme Henkelman (U Texas at Austin) reviewed latest advances in transition-state search algorithms, while Karsten Reuter (TU München) discussed the use of resulting first-principles kinetic parameters in microkinetic models and Matteo Maestri (Politecnico Milan) went up to reactor level modeling. Cynthia Friend (Harvard) complemented this catalysis focus session by highlighting present-day experimental approaches that can provide stringent references for the modeling efforts.

Wednesday's sessions then shifted towards renewable energies, with sessions on Light Harvesting and Photovoltaics. Critical parameters in this context are the energy levels of the photochromic moieties together with the transport of excited charge carriers. State-of-the-art modeling approaches, covering from time-dependent configuration interaction and density-functional theory (DFT) to semi-empirical ΔSCF-approaches, were presented in the talks by Annabella Selloni

(Princeton), Mathias Nest (TU München) and Harald Oberhofer (TU München), again complemented by an experimental overview over latest developments in organic solar cells by Clas Persson (U Oslo). In electrochemical applications, the challenges with respect to the first-principles description of charge-driven reactions are augmented by the complexities of the solid-liquid interface. Axel Groß (U Ulm) critically discussed corresponding modeling approaches under the explicit inclusion of the external bias, while Marc Koper (U Leiden) focused in the electrocatalytic context on ab initio thermodynamic approaches, in which the external potential enters indirectly in the form of a reservoir. As became quite clear in all electrochemical and photoelectrochemical talks, quantitative modeling based on first-principles is in these areas at the very beginning, and exhibits a significant time lag with respect to e.g. the explicit kinetic modeling that has been established in heterogeneous catalysis research over the last decade.

To further stimulate a perspective view away from the present-day challenges to the grand problems to be addressed in the years to come, the workshop ended with a dedicated Quo Vadis? session, in which experts from various fields were specifically asked to present their visions for the future. As acknowledged by all participants, the corresponding talks by Jens Nørskov (Stanford) and Mathieu Verstrate (U Liege) on heterogeneous catalysis and thermoelectric materials, respectively, represented a highlight of the workshop. Unfortunately, a third talk by Alan Luntz (Stanford) on battery research had to be canceled on short notice, as the speaker developed a severe neck illness.

3) Assessment of the results and impact of the event on the future directions of the field

The presented results, the talks as well as the posters, provided a comprehensive overview on the current status of the field and the next steps to take. A close contact between groups employing different methods and working in diverse application areas has been established, which was one of the main purposes of the workshop/summer school. The well selected mix of senior and junior scientists and the informal atmosphere provided room for many fruitful discussions, which allowed to assess the scope and limitations of the various approaches and to identify new important challenges for the field. The two organizers have organized many workshops and schools in the last decade(s). On this experience we can safely conclude that the scientific and personal atmosphere during this Norderney meeting was truly excellent.

Also, the feedback we received from students and senior scientists was unanimously positive. From the extended discussions it became clear how well the students had picked up the concepts, were fascinated by the different topics and started to use this to generate ideas for their own projects. The mixture of tutorials and hot topic talks was particularly commented. This achieved the right balance to illustrate on the one hand the current frontiers and its concomitant excitement, and on the other hand convey the required basic knowledge and methodology employed in this research. The students were also grateful for the possibility to present their own work in two extended poster sessions. This provided them with most valuable feedback, broke the ice and many times initiated quite detailed discussions far beyond the scope of the actual project.

The motivation for the school was that we very often note the urgent desire for dedicated training that bridges from general basics to the very methodology used

in applications in the pivotally important field of material and energy conversion at surfaces and interfaces. Seeing how well the school proceeded and how enthusiastically the students responded to the program we feel fully confirmed in our view. This school has been preceeded by a loose series of similar events, taking place every couple of years. From the success of these and the present school it is clear that a similar event will again be at place in a few years to teach the next generation of students.

Annex 4a) Programme of the meeting

Sunday, July 21, 2013

15:00 – 16:30 Registration
16:30 – 19:00 Session: Basics
Electronic Structure Theory (M. Scheffler)
Treating surfaces: Clusters vs. supercells (L. Ghiringhelli)
19:00 – 20:30 Dinner
20:45 – 22:00 Session: Surface Spectroscopy
Surface science approach, experimental techniques (M. Weinelt)

Monday, July 22, 2013

9:00 – 13:00 Session: Molecular Nanoscience
Organics at surfaces, self-assembly (Ch. Wöll)
Bonding at surfaces (A. Tkatchenko)
Hot topic 1: DFT self-interaction failures (N. Marzari)
13:00 – 15:00 Discussions & Break
15:00 – 19:00 Session: Materials for Energy
High throughput, materials genome (N. Marzari)
Scaling relations (A. Vojvodic)
Hot Topic 2: Bulk doping effects in hybrid organic/inorganic systems from quantum mechanical first principles (P. Rinke)
Hot Topic 3: Femtomagnetism: The transient band structure of Gadolinium (M. Weinelt)
19:00 – 20:30 Dinner
20:30 – 22:30 Poster Session I (even numbered posters)

Tuesday, July 23, 2013

9:00 – 13:00 Session: From Energy to Kinetics
Reactions at surfaces (C. Friend)
Barrier calculations (G. Henkelman)
Hot topic 4: First-principles based catalytic reaction engineering (M. Maestri)
13:00 – 15:00 Discussions & Break
15:00 – 19:00 Session: Heterogeneous Catalysis
Microkinetic modeling (K. Reuter)
Heat and mass transport (M. Maestri)
Hot topic 5: TBA (C. Friend)
Hot topic 6: Why (100) terraces make and break bonds (M. Koper)
19:00 – 20:30 Dinner
20:45 – 22:00 Session: Excited States
Electronic excitations, computational spectroscopy (P. Rinke)

Wednesday, July 24, 2013

9:00 – 13:00 Session: Light harvesting
Molecular levels at surfaces, optical properties (A. Selloni)
Photocatalysis at surfaces (M. Nest)
Hot topic 7: Space charge transfer at metal-oxide surfaces (M. Scheffler)

13:00 – 15:00 Discussions & Break

15:00 – 19:00 Session: Photovoltaics
Solar cells (C. Persson)
Charge transfer (H. Oberhofer)
Hot topic 8: Intriguing physics of on Cu-based solar cell materials (C. Persson)
Hot topic 9: Advances in electron dynamics with real-time time-dependent density-functional theory (M. Nest)

19:00 – 20:30 Dinner

20:30 – 22:30 Poster Session II (odd numbered posters)

Thursday, July 25, 2013

9:00 – 14:00 Session: Electrochemistry
Solid-liquid interfaces and external bias (A. Groß)
Electrocatalysis (M. Koper)
Hot topic 10: Hybrid organic/inorganic systems: Blurring borders be-tween chemisorption and physisorption (A. Tkatchenko)
Hot topic 11: Lowering reaction barriers at surfaces through concerted reaction mechanisms (A. Groß)

14:00 – 17:00 Discussions & Break

17:30 – 19:30 Conference Outing: Mudflat Hiking at the Wadden Sea

20:30 – 22:30 Dinner

Friday, July 26, 2013

9:30 – 12:30 Session: Modeling: Quo vadis?
Theory and experiment in heterogeneous catalysis (J. Nørskov)
~~Perspective on battery research (A. Luntz)~~; *canceled on short notice*
Thermoelectrics (M. Verstraete)

13:00 Departure

Annex 4b) Full list of speakers and participants

List of invited speakers:

Cynthia Friend	**Stanford (USA)**
Luca Ghiringhelli	**FHI Berlin (D)**
Axel Groß	**U Ulm (D)**
Graeme Henkelman	**U Texas (USA)**
Marc Koper	**U Leiden (NL)**
Alan Luntz	**Stanford (USA)**
Matteo Maestri	**Politecnico Milano (I)**
Nicola Marzari	**EPF Lausanne (CH)**
Mathias Nest	**TU München (D)**
Jens Nørskov	**Stanford (USA)**
Harald Oberhofer	**TU München (D)**

Clas Perrson	U Oslo (FIN)
Karsten Reuter	TU München (D)
Patrick Rinke	FHI Berlin (D)
Matthias Scheffler	FHI Berlin (D)
Annabella Selloni	Princeton (USA)
Alexandre Tkatchenko	FHI Berlin (D)
Matthieu Verstraete	U Liege (BE)
Alexandra Vojvodic	Stanford (USA)
Martin Weinelt	FU Berlin (D)
Christof Wöll	KIT Karlsruhe (D)

List of participants:

#	Familyname	Firstname
1	Abedi Khaledi	Navid
2	Abghoui	Younes
3	Ahmed Nosir	Mohamed
4	Angelova	Iva
5	Azuri	Asaf
6	Badalyan	Artavazd
7	Bonardi	Paolo
8	Bukas	Vanessa Jane
9	Carchini	Giuliano
10	Cimas	Álvaro
11	Clayborne	Andre
12	Döpper	Tibor
13	García-Muelas	Rodrigo
14	Gautier	Sarah
15	Gebhardt	Julian
16	Gobre	Vivekanand
17	Goldsmith	Bryan
18	Golze	Dorothea
19	Gossenberger	Florian
20	Graziano	Gabriella
21	Hao	Xianfeng
22	Hariharan	Seenivasan
23	Hillebrand	Philipp
24	Howard	Marco
25	Hörmann	Nicolas
26	Hussain	Javed
27	Janke	Svenja Maria
28	Josefsson	Ida
29	Kling	Tanja
30	Kozlov	Sergey
31	Ludwig	Martin
32	Maimaiti	Yasheng
33	Mancera	Luis A.

34	Müller	Moritz
35	Olimpio Pereira	Aline
36	Prucker	Veronica
37	Qi	Qi
38	Ringe	Stefan
39	Rittmeyer	Simon
40	Roman	Tanglaw
41	Ruiz Lopez	Victor Gonzalo
42	Schober	Christoph
43	Singh	Arunima
44	Sinstein	Markus
45	Son	Won-joon
46	Spickermann	Christian
47	Tonner	Ralf
48	Van den Bossche	Maxime
49	von Horsten	Frank
50	Zhao	Xunhua