Final Report

The electrode potential in electrochemistry – A challenge for electronic structure theory calculations

Castle Reisensburg near Ulm/Germany

November 26 - 29, 2017

Organizers: Axel Groß (Ulm University, Germany),
Michiel Sprik (Cambridge University, UK)

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Psi-k Network for Electronic Structure Calculations

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Processes at electrochemical electrode-electrolyte interfaces are of tremendous technological importance, in particular in the context of electrochemical energy storage and conversion. Still, atomistic details of structures and processes at these interfaces are often still not known. This calls for a close collaboration between experiment and theory on an atomistic level. However, quantum chemical studies addressing atomistic details of electrochemical interfaces face severe fundamental theoretical, computational and numerical challenges.

Among of the most severe poblems is the proper theoretical quantum chemical description of the electrode potential. In electrochemistry, structures and properties at the electrode-electrolyte interface are governed by the electrode potential which has to be kept constant along the simulation of electrochemical processes. Yet, almost all of the first-principles electronic structure studies addressing electrochemical systems are performed in the so-called constant charge mode which, however, does not correspond to the set up used in electrochemistry experiments. It was the purpose of this purely theoretical workshop, organized by Axel Groß (Ulm University, Germany) and Michiel Sprik (Cambridge University, UK), to bring together experts in the field of theoretical electrochemistry to review the current status of the field, but also to identify promising future developments. Although the main focus of the workshop was the proper theoretical description of varying electrode potentials, also other issues such as the appropriate modeling of liquid electrolytes were addressed. Besides financial support by the Psi-k Network for Electronic Structure Calculations, additional support of 13,500 EUR was provided by the German Science Foundation (DFG) which together with the contribution by Psi-k covered the expenses of the workshop.

Castle Reisensburg is an ideal location to host a workshop with about 60 participants. The participants can stay on the location which foster discussions from the morning up to late at night. Workshop announcements were posted on the internet, and already three months before the workshop started it was overbooked which confirmed that the topic of the workshop was timely. The participants including the invited speakers originated from all over the globe (see the list at the end of this report). Besides 22 participants from Germany there were 7 from Switzerland, 6 from the UK, 2 from Finland, 2 from Sweden, 2 from the Netherlands, 1 from Denmark, 1 from Ireland, 1 from Italy, 1 from France, 1 from Spain, and 1 from Slovenia. In total, 13 participants were from overseas, 10 from the USA, 1 from Canada, 1 from Japan, 1 from China. The list of participants included 45 researchers on the postdoctoral and higher level and 15 doctoral students.

As mentioned above, the talks covered various aspects of the appropriate theoretical modeling of electrochemical electrode/electrolyte interfaces. The workshop was opened by Minoru Otani, AIST Tsukuba, Japan, who had developed a first-principles constant potential scheme based on the implementation of a potentiostat in order to avoid numerical instabilities. The second opening talk was given by a rather prominent speaker, Jens Nørskov from Stanford University, USA, who described the current state and challenges with respect to the theoretical treatment of electrocatalytic reactions.

The second day was devoted to talks addressing the consideration of the electrode potential and liquid electrolytes in the description of structures and processes at electrochemical interfaces. In addition, Quiang Cui, University of Wisconsin, USA, presented a talk on equilibrium and non-equilibrium simulations of solid/liquid/biomolecule interfaces in order to broaden the scope of the workshop. Before dinner, every poster presenter had the chance to present his poster in a two-slide presentation. The disccussions in the poster session after dinner were very fruitful and intensive.

The talks on the third day were mainly devoted to the influence of the electrolyte on electrochemical processes. In order to avoid the numerically rather expensive statistical averaging over many atomic configurations, recently implicit solvent models have become rather popular. In this approach, the solvent is represented by a polarizable continuum. Opportunities, but also limitations of this approach were carefully analyzed. One of the highlights of this workshop was the presentation of Jörg Neugebauer, MPI for Iron Research, Düsseldorf, who presented a a novel approach that allows to run molecular dynamics simulations of reactions at electrodes under controlled potentiostat conditions and that can be easily implemented in standard DFT codes. The last two talks were given by two young researchers whose presentations were selected from the contributed talks. The third day ended with the conference dinner which provided a nice framework for the ongoing discussions about the appropriate theoretical description of electrochemical interfaces.

The last day started with two invited talks, followed again by two talks of young researchers, before the workshop was wrapped up by the two organizers. There are certainly still many challenges associated with the theoretical modeling of electrochemical interfaces. The field of atomistic theoretical electrochemistry has not matured yet, as many different theoretical, computational and numerical techniques are applied, and there is still a significant need for the development of reliable theoretical methods and algorithms. Still, some promising approaches have been presented in this workshop. In particular, it has now been generally accepted that the proper consideration of the electrochemical environment is crucial in order to reliably describe processes and structures at these interfaces.

This field is also particularly interesting for young researchers, as reflected by the large number of 15 doctoral students who registered for the workshop. It combines challenging scientific, theoretical and numerical questions with a high societal relevance with respect to the electrochemical energy storage and conversion which will be a cornerstone of our future energy technology. The format of the workshop providing ample time for discussions at a stimulating location allowed intensive interactions between junior and senior scientists. Besides the review of the current status of the field and the identification of promising future developments we consider numerous and active participation of young researchers starting from the doctoral level as one of the main achievements of the workshop.

Meeting programme

Schedule Sunday, November 26, 2017

14:00 - 18:30	*Arrival and registration*	
18:30 - 20:00	*Dinner*	
20:00 - 20:15	Axel Groß, Michiel Sprik	*Introduction*
20:15 - 21:00	Minoru Otani	*Electrochemical reaction simulations at metal/liquid interfaces using effective screening medium + implicit classical solvation methods*
21:00 - 21:45	Jens Nørskov	*Describing catalytic reactions at the electrified solid-liquid interface*

Monday, November 27, 2017

08:00 - 09:00	Breakfast	
09:00 - 09:45	Jean-Sebastian Filhol	*Insights into electrochemical modelling and effects at interfaces*
09:45 - 10:30	Karen Chan	*Modelling the electrochemical interface: applications to* CO_2 *reduction*
10:30 - 11:00	Coffee break	
11:00 - 11:45	Clotilde Cucinotta	Pt-*water double layer structure in realistic solution conditions*
11:45 - 12:30	Qiang Cui	*Equilibrium and non-equilibrium simulations of solid/liquid/biomolecule interfaces*
12:30 - 14:30	Lunch	
14:30 - 15:15	Harald Oberhofer	*Modelling photo-electrochemistry beyond the computational hydrogen electrode*
15:15 - 16:00	Karsten Reuter	*Implicit Solvation Methods in the FHI-aims Code*
16:00 - 16:30	Coffee break	
16:30 - 18:00		*Short presentation of posters*
18:00 - 20:00	Dinner	
20:00 - 22:00	*Poster Session*	

Tuesday, November 28, 2017

08:00 - 09:00	Breakfast	
09:00 - 09:45	Oliviero Andreussi	*Continuum approaches in electrochemistry: methods, and applications of ENVIRON*
09:45 - 10:30	Johannes Lischner	*Ab initio electrochemistry with joint density-functional theory*
10:30 - 11:00	Coffee break	
11:00 - 11:45	Jan Rossmeisl	*Ions Influence on Electrocatalysis*
11:45 - 12:30	Jun Cheng	*Simulating electrochemical interfaces using DFTMD*
12:30 - 14:30	Lunch	
14:30 - 15:15	Mira Todorova	*Selective stabilization of polar oxide surfaces in electrochemical environment*
15:15 - 16:00	Jörg Neugebauer	*A first principles approach to model electrochemical reactions in an electrolytic cell*
16:00 - 16:30	Coffee break	
16:30 - 17:15	Alfredo Pasquarello	*Electron Energy Levels in Liquid Water and at Semiconductor-Water Interfaces*
17:15 - 17:45	Ho Viet Thang	*DFT Study of Electron Transfer at* ZnO/X(111) *Interfaces* (X=Cu, Ag, Au)
17:45 - 18:15	Jinggang Lan	*Insights into the interface* CO/Pt(111) *with water from First-Principles Simulations*
18:30 - 21:00	Conference Dinner	
21:00 -	Social Event	

Wednesday, November 29, 2017

08:00 - 09:00	Breakfast	
09:00 - 09:45	Michael Eikerling	*Deciphering the Oxygen Reduction Reaction with Theory and Computation*
09:45 - 10:30	Wolfgang Schmickler	*Charge and Potential in Nanotubes and -slits*
10:30 - 11:00	Coffee break	
11:00 - 11:30	Sung Sakong	*Thermodynamic description of the pH dependence of the potential of zero charge (pzc) on a* Pt(111) *electrode*
11:30 - 12:00	Ian McCrum	*Approximating the effects of solvent and electric field: Understanding the pH dependence of* H/OH *adsorption on* Pt
12:00 - 12:15	Axel Groß, Michiel Sprik	*Concluding remarks*
12:15 - 13:30	Lunch	
13:30	Departure	

List of participants

	Name	Affiliation	Role
1	Dr. Oliviero Andreussi	EPFL Lausanne, Switzerland	Speaker
2	Dr. Lars Blumenthal	Imperial College London, UK	Poster Presenter
3	Dr. Assil Bouzid	EPFL Lausanne, Switzerland	Poster Presenter
4	Dr. Vanessa Bukas	Stanford University, USA	Poster Presenter
5	Dr. Miguel Caro	Aalto University, Finland	Poster Presenter
6	Arcesio Castaneda Medina	Ruhr University Bochum	Poster Presenter
7	Dr. Karen Chan	Stanford University, USA	Speaker
8	Dr. Leanne D. Chen	CalTech, Los Angeles, USA	Poster Presenter
9	Prof. Dr. Jun Cheng	Xiamen University, China	Speaker
10	Dr. Stephen J. Cox	Cambridge University, UK	Poster Presenter
11	Dr. Clotilde Cucinotta	Trinity College Dublin, Ireland	Speaker
12	Prof. Dr. Quiang Cui	Univ. of Wisconsin, USA	Speaker
13	Colin Dickens	Stanford University, USA	Poster Presenter
14	Dr. Katharina Doblhoff-Dier	Leiden University, Netherlands	Participant
15	Prof. Dr. Michael Eikerling	Simon Fraser Univ., Canada	Speaker
16	Dr. Holger Euchner	Helmholtz Institute Ulm	Participant
17	Meredith Fields	Stanford University, USA	Poster Presenter
18	Prof. Dr. Jean-Sebastian Filhol	Universite Montpellier, France	Speaker
19	Dr. Guiseppe Fisicaro	University of Basel, Switzerland	Poster Presenter
20	Dr. Zdenek Futera	University College London, UK	Poster Presenter
21	Dr. Aleksej Goduljan	Ulm University	Participant
22	Prof. Dr. Axel Groß	Ulm University	Organizer
23	Dr. Svante Heström	Stockholm University, Sweden	Poster Presenter
24	Maximilian Hödl	MPI-FKF Stuttgart	Poster Presenter
25	Dr. Nicolas Hörmann	EPFL Lausanne, Switzerland	Speaker
26	Prof. Dr. Marcella Ianuzzi	University Zurich, Switzerland	Participant
27	Markus Jäckle	Helmholtz Institute Ulm	Participant
28	Dr. Fernanda Juarez	Ulm University	Participant
29	Charlotte Kirk	Stanford University, USA	Poster Presenter
30	Dr. Manuel Kolb	Stockholm University, Sweden	Poster Presenter
31	Anja Kopac Lautar	University of Ljubljana, Slovenia	Poster Presenter
32	Jinggang Lan	University Zurich, Switzerland	Poster Presenter
33	Dr. Johannes Lischner	Imperial College London, UK	Poster Presenter
34	Dr. Matthias May	Cambridge University, UK	Poster Presenter
35	Dr. Ian McCrum	Leiden University, Netherlands	Speaker
36	Dr. Markus Melander	University of Jyväskylä, Finland	Poster Presenter
37	Dr. Maryam Naderian	Ulm University	Participant
38	Prof. Dr. Jörg Neugebauer	MPI Düsseldorf, Germany	Speaker
39	Dr. Huu Chuong Nguyën	ICIQ Barcelona, Spain	Poster Presenter
40	Prof. Dr. Jens Nørskov	Stanford University, USA	Speaker
41	Dr. Harald Oberhofer	TU Munich, Germany	Speaker
42	Daniel Opalka	TU Munich, Germany	Participant
43	Prof. Dr. Minoru Otani	AIST Tsukuba, Japan	Speaker
44	Dennis Pache	Ruhr University Bochum	Poster Presenter

	Name	Affiliation	Role
45	Prof. Dr. Alfredo Pasquarello	EPFL Lausanne, Switzerland	Speaker
46	Prof. Dr. Karsten Reuter	TU Munich, Germany	Speaker
47	Brian Rohr	Stanford University, USA	Poster Presenter
48	Prof. Dr. Jan Rossmeisl	Univ. of Copenhagen, Denmark	Speaker
49	Dr. Sung Sakong	Ulm University	Speaker
50	Hannah Schlott	University of Erlangen	Poster Presenter
51	Prof. Dr. Wolfgang Schmickler	Ulm University	Speaker
52	Dr. Rochus Schmid	Ruhr University Bochum	Poster Presenter
53	Aayush Singh	Stanford University, USA	Poster Presenter
54	Prof. Dr. Eckhard Spohr	Uni Duisburg-Essen	Participant
55	Prof. Dr. Michiel Sprik	Cambridge University, UK	Organizator
56	Sudarsan Surendaral	MPI Düsseldorf, Germany	Poster Presenter
57	Dr. Ho Viet Thang	Univ. of Milano-Bicocca, Italy	Speaker
58	Dr. Mira Todorova	MPI Düsseldorf, Germany	Speaker
59	Dr. Stefan Wippermann	MPI Düsseldorf, Germany	Poster Presenter
60	Suhyun Yoo	MPI Düsseldorf, Germany	Poster Presenter

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