“Modelling materials and chemistry – in an eSSENCE and a European context”

Kersti Hermansson, Department of Chemistry-Ångström, UU
kersti@kemi.uu.se

Swedish e-science Academy 2015, 14-15 October 2015
"Chemistry of complex materials"
– a brief exposé of our eSSENCE-enabled achievements
"Chemistry of complex materials"
– a brief exposé of our eSSENCE-enabled achievements

New eSSENCE recruitment:
bitr. lektor
Peter Broqvist
"Chemistry of complex materials"
– a brief exposé of our eSSENCE-enabled achievements

New eSSENCE recruitment
(bitr. lektor)
Peter Broqvist

New methods
Modelling materials and chemistry

This is a challenge because...


* Short & long range interactions, strong & weak interactions at the same time!

* Dynamics (temperature) is often crucial to incorporate!

* Large systems and long times with adequate accuracy are impossible/difficult to compute.
Example 1

"Multiscale modelling"

Method/model development is needed both at each level and between levels (method integration).
Our computational strategy for chemistry in and on materials

Broqvist, Kullgren, Rafieefar, Hermansson, ongoing work.
in collaborations with the main developers of ReaxFF (van Duin) and DFTB+ (Frauenheim)
Multiscale simulation environment

Based on ASE*
(Atomistic simulation environment)
- Easy to use
- Flexible
- Customizable
- Pythonic

Reactive Force Field

Parameterization

Approximate DFT

QM

*https://wiki.fysik.dtu.dk/ase/
Getting the DFTB parameters

Generate Slater-Koster tables

Compare:
- Band structure
- $E_{\text{electronic}}$

Sufficient quality?

No

Choose/adjust:
- Wavefunction compression
- Potential compression

Yes

Fit repulsive potential

Compare:
- Surface energies
- Vacancy formation energies
- Local structure and relaxation

Sufficient quality?

No

Choose/adjust:
- Hubbard $U$
- $\beta$-occupancy

Yes

FINAL PARAMETERS
Example 2

**Embedded-cluster model**

*Step 1. Calculation of the reference electrostatic potential*

*Step 2: Optimization of the positions and magnitudes of the embedding PCs.*

*Step 3: Validation at the testing grid points*

Our embedded-cluster models consist of four layers:

- Cluster of quantum mechanically treated atoms
- Surrounded by large core Ce-ECPs without basis sets
- Shell-model point charges at the positions of Ce and O ions
- A sphere of fitted point charges which reproduces the electrostatic potential from the remaining surface
Embedded-cluster results for CO adsorbed on top of a CeO$_2$ surface

QM cluster seen from above. The embedding is not shown.

Embedding model

Adsorption energy on top of Ce

<table>
<thead>
<tr>
<th>Method</th>
<th>Energy (kJ/mol)</th>
</tr>
</thead>
<tbody>
<tr>
<td>HF</td>
<td>15</td>
</tr>
<tr>
<td>MP2, full</td>
<td>24</td>
</tr>
<tr>
<td>MP2, incr.</td>
<td>24</td>
</tr>
<tr>
<td>CCSD(T), incr.</td>
<td>21</td>
</tr>
</tbody>
</table>

Timing estimates:

<table>
<thead>
<tr>
<th>Method</th>
<th>Time</th>
</tr>
</thead>
<tbody>
<tr>
<td>HF</td>
<td>17 hours</td>
</tr>
<tr>
<td>MP2, full</td>
<td>8 days</td>
</tr>
<tr>
<td>MP2, incr.</td>
<td>3 days</td>
</tr>
<tr>
<td>CCSD(T), full</td>
<td>est. 35 years</td>
</tr>
<tr>
<td>CCSD(T), incr.</td>
<td>23 days</td>
</tr>
</tbody>
</table>
Embedded-cluster vs. periodic model

3-D periodicity

2-D periodicity

In both cases:

BUT: Non-periodic

2 advantages!
Example 3

\( \text{QM}_{\text{el}} + \text{MD} + \text{QM}_{\text{el}} + \text{QM}_{\text{nuc}} \)

![Diagram showing QM cluster calculations, MD simulations, and OH spectrum analysis](image-url)
"Chemistry of complex materials"
– a brief exposé of our eSSENCE-enabled achievements

New methods

New eSSENCE recruitment (bitr. lektor)

New results
Example 1.

DFTB performance for nanoparticles

Peter Broqvist et al. To be published
Example 2. Calculations meet experiments

SEM image of ceria (CeO$_2$) nanocrystals

"SEM image of ceria (CeO$_2$) octahedra"
[Z. Wu et al., Journal of Catalysis 285, 61 (2012)].

We discovered the mechanism


It was found experimentally 2012 that the oxygen storage capacity (cf. car catalysts) of very small CeO$_2$ nanoparticles increases drastically. O$_2^-$ molecules were found, but no mechanism.
Example 3. Calculations meet experiments

Conclusion: The agreement is almost perfect! So then we can make use of all the extra mechanistic detail provided by the calculations!
"Chemistry of complex materials"
– a brief exposé of our eSSENCE-enabled achievements

New eSSENCE recruitment (bitr. lektor)

New methods

New results

National initiative III.

CECAM

National initiative I.
Multi-scale modelling meeting

National initiative II.
VR-infra
Another activity on the national arena: discussion of "Swedish science cases for e-infrastructure"

Chapter authors: Profs. Henrik Grönbeck, Erik Lindahl, Igor Abrikosov, Kersti Hermansson

7 — Materials, Chemistry and Nano-science

- Materials, Chemistry and Nano-science constitute the largest community of HPC users in Sweden and worldwide.
- There is a need for an increase in available computer power of orders of magnitude.
- It will become possible to move into a paradigm of predictive simulation of materials and molecular design.
- New understanding will emerge from large-scale simulations of materials, nano-systems and chemical phenomena.
- Accurate and realistic calculations of complex and dynamic systems will become possible.
- There is a need for development of methods, models and software.
- There is a need for application experts at different levels.
Green's function methods: the next generation II
Location: CECAM-HQ-EPFL, Lausanne, Switzerland
May 4, 2015 - May 7, 2015
- Arjan Berger
  University Paul Sabatier, France
- Pina Romaniello
  University Paul Sabatier, France
- Francesco Sotile
  Ecole Polytechnique, Palaiseau, France

Molecular hydrodynamics meets fluctuating hydrodynamics.
Location: CECAM-ES, Residencia La Cristalera, Miraflores de la Sierra, Madrid.
May 10, 2015 - May 14, 2015
- Ignacio Pagonabarraga Mora
  University of Barcelona, Spain
- Rafael Delgado-Buscalioni
  Autonomous University of Madrid, Spain
- Pep Español
  National University of Distance Education, Spain

From Many-Body Hamiltonians to Machine Learning and Back
Location: CECAM-DE-MM1P, Dahlem, Berlin, Germany
May 11, 2015 - May 13, 2015
- Matthias Rupp
  University of Basel, Switzerland
- Alexandre Tkatchenko
  Fritz Haber Institute of the Max Planck Society (FHI), Berlin, Germany
Development of next generation accurate approximate DFT/B methods - Flagship workshop and tutorial
Location: CECAM-DE-MM1P, University of Bremen, Germany
October 11, 2015 - October 15, 2015
- Thomas Frauenheim
  University of Bremen, BCCMS, Germany
- Fernand Spiegelman
  CNRS and Université de Toulouse, France
- Marcus Elstner
  Karlsruhe Institute of Technology, Germany

Free energy landscapes for protein folding. Consensus or dissensus?
Location: Building HIT room E 51 ETHZ, Zurich, Switzerland
October 12, 2015 - October 14, 2015
- Emanuele Paci
  University of Leeds, United Kingdom
- Sergei Krivov
  Leeds University, United Kingdom
- Amedeo Caflisch
  University of Zurich, Switzerland

A scientific roadmap for simulation and modelling for 2020
Location: CECAM USI-Lugano, Switzerland
September 16, 2015 - September 18, 2015
- Dominic Tildesley
  Swiss Federal Institute of Technology Lausanne (EPFL), Switzerland
- Sara Bonella
  CECAM@EPFL, Switzerland
"Chemistry of complex materials"
– a brief exposé of our eSSENCE-enabled achievements

- New methods
- New eSSENCE recruitment (bitr. lektor) Peter Broqvist
- International initiative II.
- The European Materials Modelling Council
- National initiative I.
- National initiative II.
- VR-infra
Vision for model & method development

European materials-based industries need to have access to models and modelling workflows of a high enough quality, reliability and predictive power such that modelling results will make an economical, environmental or societal difference. Therefore it is important for this WG to, on the one hand,

- **promote** the improved and wider exploitation of *existing* models and methods, and on the other,

- **promote** the development of *new, more reliable* (yet computationally feasible) models and methods.

Biggest challenges in model & method development

- Accuracy - predictability
- Complexity => towards realistic systems
- Enhance the role of discrete models in industrial modelling.
- Communication - between models (multiscale modelling, C/L, materials informatics, interoperability, ...)
  - between people
"130 High profile speakers and more than 1000 delegates from private and public organizations, investors, enterprises, SMEs and national and European policy makers will merge industrial leadership and societal challenges into a new vision."

**Future for modelling: materials and industrial process**

**Speakers:**
- Nicola Marzari        EPFL, Lausanne
- Seiji Kajita              Toyota Central R&D Labs., Inc
- Carlo Cavazzoni      CINECA Supercomputer Center
- Peter Klein               Fraunhofer ITWM
- Alain Bernard          IRT Jules Verne, Ecole Centrale de Nantes

**Chairman and moderator:**
- Kersti Hermansson  Uppsala University, Sweden
New eSSENCE recruitment
(bitr. lektor)
Peter Broqvist

New methods

New results

National & international initiative
CECAM

National initiative
I.
Multi-scale modelling meeting

National initiative
II.
VR-infra

+ Nice contacts and people!!