
**Title:** Electronic Structure Theory with Numeric Atom-Centered Basis Functions

**Location:** Modelling Center TUM.Computational, Technical University of Munich

**Sponsors:** Psi-K and MS1P (Molecular Dynamics from First Principles)

**Dates:** 9 July 2018 to 11 July 2018

**Organizers**: Volker Blum (Duke University), William Huhn (Duke University), Harald Oberhofer (Technical University of Munich), Xinguo Ren (University of Science and Technology of China), Mariana Rossi (Fritz Haber Institute of the Max Planck Society)

## State of the Art

Electronic structure theory (EST) including density-functional, wave-function, and many-body perturbation theory based methods continues to grow in reach and importance across computational molecular and materials science. For non-periodic and periodic models up to thousands of atoms in size, this first- principles, i.e., quantum mechanics based approach offers unbiased predictions and understanding that meets basic science, materials engineering, and industrial needs. Application examples include electronic materials, energy related materials for photovoltaics or photocatalysis, biomolecular phenomena and reactions, etc. A key methodological choice that distinguishes different successful implementations of electronic structure theory is the choice of the quantum-mechanical basis set used to discretize the underlying mathematical equations. This workshop focuses on new developments that leverage localized, numeric atom-centered orbital (NAO) basis functions, a choice upon which a number of the strongest available electronic structure developments are founded. Examples include the Siesta code [1], the FPLO code [2], DMol3 [3], ADF [4], PLATO [5], the FHI-aims code [6], and many others. In addition, key concepts in NAO codes are closely related to formalisms employed with other types of basis sets, such as Gaussian-type orbitals, linearized augmented

plane waves, etc. Specific advantages of NAO-based electronic structure codes include the fairly straightforward modeling of non-periodic and periodic systems on equal footing, suitability for order-N scaling numerical algorithms [7], efficient scaling towards large systems [8,9], efficient algorithms for exact exchange and many-body approaches based on the two-electron Coulomb operator [10,11], and the proven potential for simulations from fast qualitative models up to high-accuracy calculations that match the best available benchmark codes [12,13,14,15].

Among the drivers for advances in NAO-based electronic structure theory in general, the community built around the FHI-aims code plays an important role. This is, by now, a large community, with key strengths in high-accuracy simulations, scalability to large systems, approaches beyond "simple" density functional theory, and a deep involvement in several other community-wide software and infrastructure efforts. This development continues at high pace, with recent examples (in the past two years) including:

- the ELectronic Structure Infrastructure (ELSI), which provides an interface layer between DFT codes and methods that solve or circumvent the Kohn-Sham eigenvalue problem to enable massively- parallel, large-scale DFT [16]
- a Density Functional Perturbation Theory implementation based on NAO basis functions which is efficient and gives easy access to spatially localized quantities [17]
- an efficient implementation of spin-orbit coupling usable for large-scale hybrid-functional calculations, coupled with a broad validation of relativistic effects across the periodic table [14]
- a highly efficient implicit solvation scheme, including ionic effects with both finite ion sizes and an electrolytic Stern layer, based on the size-modified Poisson-Boltzmann equation and a multipole moment relaxation (MERM) solver [18]
- a state of the art fragment orbital method for determining electronic couplings between approximate molecular diabatic states for use in e.g. high throughput screening of organic semiconductors [19]

and many other small and large methodological developments in electronic structure theory. Importantly, these developments do not occur in isolation, but instead build heavily on continuing interaction, collaboration, and feedback from the wider community, e.g., involvement in the development of solver libraries, the “Delta test” community benchmark in electronic structure theory [12], implementation of advanced exchange-correlation functionals, the ASE environment, the i-PI program package [20], “big data” efforts such as NoMaD, the CECAM sponsored ESL effort, and others.

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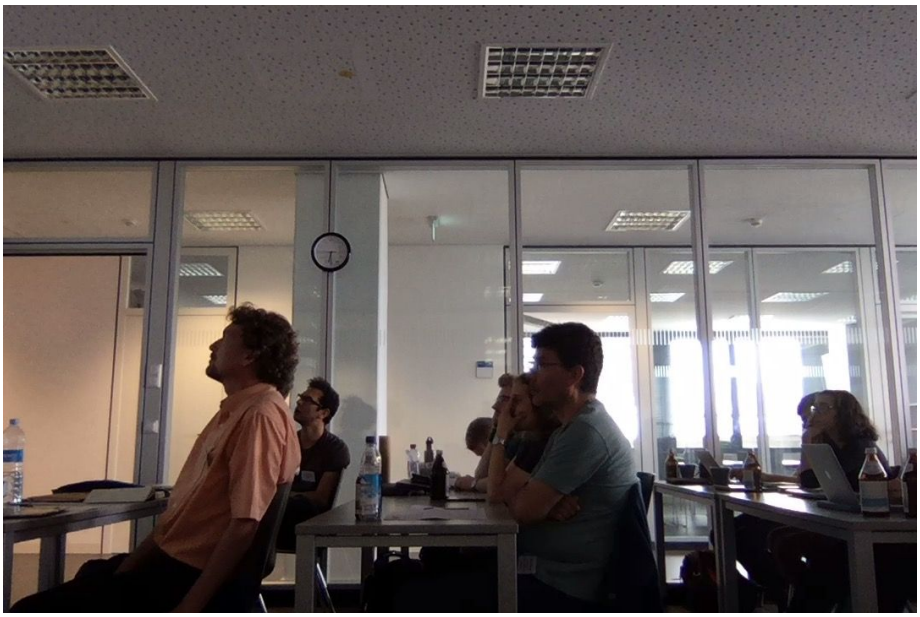
## Description and Outcomes



This workshop focused on methods that leverage localized, numeric atom-centered orbital (NAO) basis functions, a choice upon which a number of the strongest available electronic structure developments are founded. The workshop brought together key players from the FHI-aims code and related European and international efforts to highlight, discuss, and advance the state of the art of NAO-based modeling of molecules and materials based on the first principles of quantum mechanics.

This workshop covered three days and 23 invited talks, covering:

- development of community-based, shared infrastructure projects for electronic structure theory (Garcia, Larsen, Pouillon),
- benchmarking efforts to assess and improve the accuracy of approximations used in electronic structure theory (Al-Hamdani, Goedecker, Liu),
- applications of density functional perturbation theory (Laasner, Raimbault, Shang),
- automation of workflow via machine learning and “big data” efforts (Ghiringhelli, Hoja),
- scalability towards large systems and exascale computational resources (Huhn, Scheurer, Yu),

- numerical algorithms and new methods for NAO-based electronic structure theory (Hermann, Ringe, Rossi), and
- extensions beyond standard Kohn-Sham DFT (Golze, Havu, Michelitsch, Oberhofer, Ren)

A poster session with 14 posters contributed by workshop participants was held on the first night.

Afternoons and evenings were reserved for "Hands-On Discussions", in which participants split up into small groups to focus on topics of specific interest ranging from methodological improvements all the way to code development questions related to specific subgroups of participants and joint programming sessions. A list of topics that were addressed during these hands-on sessions included:

- Tutorial sessions for various EST and computational packages:
  - the massively parallel genetic algorithm code GAtor
  - the build system generator CMake
  - the Atomic Simulation Environment
  - the Electron Structure Library software bundle
- Method development:
  - improving convergence of the SCF cycle,
  - modeling van der Waals interactions for real materials,
- Code development:
  - code development best practices for distributed software projects
  - maintaining test suites to ensure binary reliability,
  - controlling dependencies to ensure compilation across multiple platforms

## Community Needs

One consistent theme that emerged throughout the discussion session was the growing complexity of software development practices. It was critical to have shareholders from various software projects (FHI-aims, ELSI, ASE, Abinit, SIESTA, GAtor, the ESL Bundle) to share their strategies for ensuring maintainability of their code base.

- Proper versioning for libraries
- Consistent testing strategy
- Collaboration across "competing" projects to address software issues

## Workshop Attendees

Experienced Participants (with Ph. D.'s)

| | Surname | Given Name | Institution | Country |
|---|---|---|---|---|
| 1 | **Al-Hamdani** | **Yasmine** | University of Luxembourg | Luxembourg |
| 2 | **Blum** | **Volker** | Duke University | USA |
| 3 | **Carbogno** | **Christian** | Fritz Haber Institute of the Max Planck Society | Germany |
| 4 | **Garcia** | **Alberto** | Institute of Materials Science of Barcelona | Spain |
| 5 | **Ghiringhelli** | **Luca** | Fritz Haber Institute of the Max Planck Society | Germany |
| 6 | **Goedecker** | **Stefan** | University of Basel | Switzerland |
| 7 | **Havu** | **Ville** | Aalto University | Finland |
| 8 | **Huhn** | **William** | Duke University | USA |
| 9 | **Laasner** | **Raul** | Duke University | USA |
| 10 | **Larsen** | **Ask** | University of the Basque Country | Spain |
| 11 | **Oberhofer** | **Harald** | Technical University of Munich | Germany |
| 12 | **Pouillon** | **Yann** | University of the Basque Country | Spain |
| 13 | **Raimbault** | **Nathaniel** | Fritz Haber Institute of the Max Planck Society | Germany |
| 14 | **Ren** | **Xinguo** | University of Science and Technology of China | China |
| 15 | **Reuter** | **Karsten** | Technical University of Munich | Germany |
| 16 | **Ringe** | **Stefan** | Stanford University | USA |
| 17 | **Rodriguez-Fortea** | **Antonio** | Rovira i Virgili University | Spain |
| 18 | **Rossi** | **Mariana** | Fritz Haber Institute of the Max Planck Society | Germany |
| 19 | **Scheffler** | **Matthias** | Fritz Haber Institute of the Max Planck Society | Germany |
| 20 | **Scheurer** | **Christoph** | Technical University of Munich | Germany |
| 21 | **Shang** | **Honghui** | Fritz Haber Institute of the Max Planck Society | Germany |
| 22 | **Wang** | **Haiyuan** | Fritz Haber Institute of the Max Planck Society | Germany |

PhD Students

| | Surname | Given Name | Institution | Country |
|---|---|---|---|---|
| 23 | **Altschäffel** | **Jan** | University of Göttingen | Germany |
| 24 | **Arefi** | **Hadi** | Peter Grünberg Institute | Germany |
| 25 | **Bi** | **Sheng** | Fritz Haber Institute of the Max Planck Society | Germany |
| 26 | **Bier** | **Imanuel** | Carnegie Mellon University | USA |
| 27 | **Erker** | **Simon** | Graz University of Technology | Austria |

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| 28 | **Filser** | **Jakob** | Technical University of Munich | Germany |
| 29 | **Golze** | **Dorothea** | Aalto University | Finland |
| 30 | **Hekele** | **Joscha** | University of Duisburg-Essen | Germany |
| 31 | **Hermann** | **Jan** | Fritz Haber Institute of the Max Planck Society | Germany |
| 32 | **Hoja** | **Johannes** | University of Luxembourg | Luxembourg |
| 33 | **Karcz** | **Adam** | Technical University of Denmark | Denmark |
| 34 | **Kick** | **Matthias** | Technical University of Munich | Germany |
| 35 | **Knoop** | **Florian** | Fritz Haber Institute of the Max Planck Society | Germany |
| 36 | **Lenz** | **Maja-Olivia** | Fritz Haber Institute of the Max Planck Society | Germany |
| 37 | **Liu** | **Chi** | Duke University | USA |
| 38 | **Michelitsch** | **Georg** | Technical University of Munich | Germany |
| 39 | **Moreno Vicente** | **Antonio** | Rovira i Virgili University | Spain |
| 40 | **Muschielok** | **Christoph** | Technical University of Munich | Germany |
| 41 | **Timmermann** | **Jakob** | Technical University of Munich | Germany |
| 42 | **Wagner** | **Frank** | Max Planck Institute for Chemical Physics of Solids | Germany |
| 43 | **Yu** | **Victor** | Duke University | USA |

## Workshop Schedule

# Day 1 (Mon, July 9)

## Opening

**08:30-09:00** Arrival and Registration
**09:00-09:30** Welcoming Remarks
**09:30-10:00** Volker Blum (Duke)
*State of FHI-aims*

## Morning Session

**10:00-10:30** Yann Pouillon (Basque Country)
*The Electronic Structure Library: Community-Driven Development of Software Libraries for Electronic Structure Simulations*
**10:30-11:00** Coffee Break
**11:00-11:30** Alberto García (ICMAB)
*Advances in SIESTA: New Functionalities, Domain Libraries, and Scripting Interfaces*
**11:30-12:00** Ask Larsen (Basque Country)
*ASE: Overview and Developments*

**12:00-13:30** Lunch

## Afternoon Session

**13:30-14:00** Stefan Goedecker (Basel)
*Exact DFT Results and Accuracy Benchmarking of Atom Centered Basis Sets and Pseudopotentials*
**14:00-14:30** Yasmine Al-Hamdani (Luxembourg)
*Assessing XC Functionals for Non-Covalently Interacting Materials using Diffusion Monte Carlo*
**14:30-15:00** Chi Liu (Duke)
*Benchmark of Ab Initio Bethe-Salpeter Equation Approach with Numeric Atom-Centered Orbitals*
**15:00-15:30** Xinguo Ren (USTC Hefei)
*Periodic GW with Numeric Atom-Centered Basis Functions: Recent Progress and Challenges*
**15:30-16:00** Coffee Break

## Evening

**16:00-18:00** Hands-On Discussions (1)
**18:00-21:00** Conference Dinner at Aumeister Restaurant und Biergarten im Englischen Garten

# Day 2 (Tues, July 10)

## Morning Session

**09:30-10:00** Nathaniel Raimbault (FHI Berlin)
*Raman Spectra of Molecular Crystals from DFPT*
**10:00-10:30** Coffee Break
**10:30-11:00** Raul Laasner (Duke)
*Molecular NMR Shieldings, J-Couplings, and Magnetizabilities from Numeric Atom-Centered Orbital Based Density-Functional Theory*
**11:00-11:30** Luca Ghiringhelli (FHI Berlin)
*Data-Driven Materials' Properties Maps: Methods and Infrastructure*
**11:30-12:00** Johannes Hoja (Luxembourg)
*Reliable and Practical Computational Prediction of Molecular Crystal Polymorphs*
**12:00-13:30** Lunch

## Afternoon Session

**13:30-14:00** William Huhn (Duke)
*GPU-Accelerated Real Space Electronic Structure Theory on HPC Resources*
**14:00-14:30** Christoph Scheurer (TU Munich)
*Update on the ELPA-AEO project*
**14:30-15:00** Victor Yu (Duke)
*Open Infrastructure for Large-Scale Kohn-Sham Density-Functional Theory: The ELSI Project*
**15:00-15:30** Mariana Rossi (FHI Berlin)

*Approximations to Quantum Dynamics in Weakly Bonded Systems Through the FHI-aims/i-PI Interface*
**15:30-16:00** Coffee Break

## Evening

**16:00-18:00** Hands-On Discussions (2)
**18:00-20:00** Poster Session with Light Dinner

# Day 3 (Wed, July 11)

## Morning Session

**09:00-09:30** Stefan Ringe (Stanford)
*Continuum Solvent Modeling in FHI-aims: Current State of the SMPB Method*
**09:30-10:00** Georg Michelitsch (TU Munich)
*Current State in the Simulation of Core-Level Spectroscopies in FHI-aims*
**10:00-10:30** Honghui Shang (FHI Berlin)
*Density-Functional Perturbation Theory in FHI-aims*
**10:30-10:45** Coffee Break
**10:45-11:15** Jan Hermann (FHI Berlin)
*Modeling van der Waals Interactions in Materials with Many-Body Dispersion*
**11:15-11:45** Ville Havu (Aalto)
*Surface Stoichiometry of Cu(In,Ga)Se2 Solar-Cell Absorber: DFT and HAXPES Study*
**11:45-12:15** Harald Oberhofer (TU Munich)
*Embedding Quantum Regions in Classical Environments with FHI-aims*
**12:15-13:30** Lunch

## Afternoon Session

**13:30-14:00** Dorothea Golze (Aalto)
*Accurate Core-Level Spectra from GW*
**14:00-14:30** Wrap-Up Session and End of Workshop