Report on
" Quo vadis Self-Interaction Correction (QVSIC)?
September 23–26 2019, Freiberg, Germany"

June 26, 2020

Details of the workshop

- **Webpages:**

  - Workshop:
    `https://tu-freiberg.de/en/fakultaet2/thph/tagungen/sic-2019`
    `https://indico.physik.tu-freiberg.de/event/1/`
  - Method:
    `https://flosic.physik.tu-freiberg.de/`
  - Code:
    `https://github.com/pyflosic`

- **Organizers:** Prof. Jens Kortus and Prof. Koblar Alan Jackson
- **Local organizer:** Sebastian Schwalbe
- **Funding and support:** Psi-K, Technical University Bergakademie Freiberg,
  Central Michigan University

Figure 1: QVSIC group picture. Prof. John P. Perdew and Prof. Hannes Jónsson are in the middle of the first row.

Motivation

The main topics were:

- Self-interaction correction (SIC)
- Successes and failures of SIC
- Real and complex-valued orbitals for SIC
- Generalizations of SIC
- Alternative SIC methods

The workshop *Quo vadis Self-Interaction Correction (QVSIC)?* brought together together senior experts and young researchers from all over the world. The workshop took place at *Deutsches Brennstoffinstitut* in Freiberg, Germany and 27 scientists from 6 countries attended. The participation of Prof. John P. Perdew, Prof. Hannes Jónsson and Prof. Mark R. Pederson, three researchers who have developed important concepts of self-interaction correction (SIC) within density functional theory (DFT) was a great experience and an honour for all the participants.

The scientific program contained 3 plenary talks, 15 invited talks and 10 poster presentations. Further details of the workshop can be found online (see

details of workshop). As many participants provided insights into unpublished work, we decided to provide only the respective titles of the presentations along with the presenter names.

After the scientific sessions, the last day of the workshop offered a hands-on session for Ph.D students and postdocs introducing the PyFLOSIC code. This possibility has been used by 9 scientists. In addition, other SIC implementations were discussed, compared, and partially tested, too. During the workshop, 3 extensive open roundtables enabled fruitful discussions, which helped to clarify the current status of SIC and showing possible ways to improve the method further.

Monday

The first day of the workshop started with a welcoming speech by the organizers. The day continued with plenary talks by Prof. John P. Perdew and Prof. Hannes Jónsson. This opening session provided a profound introduction to the topic of the workshop discussing successes as well as problems of current methods to treat self-interaction error.

Prof. Perdew explained the core ideas of a proper SIC within DFT and how to apply SIC to higher rungs of density functional approximations. The differences between functional development based on fulfilling appropriate constraints and norms and development based on fitting functionals to datasets were discussed. Prof. Perdew also presented recent results that demonstrate that SIC fails to give correct results in the limit of a slowly-varying density. It thus breaks norms that are satisfied by the uncorrected density functional methods. Prof. Perdew gave further insights into the importance of noded or lobed one-particle densities within real and complex-valued SIC. In addition, possible improvements of SIC like global or local scaling ideas were introduced to the audience.

Prof. Jónsson pointed out the importance of complex orbitals and how a proper scaling can improve standard SIC results. Many selected examples like CH_4, the CH_3CN^- dipole-bound anion, Rydberg states in trimethylamine (TMA), the delocalized hole in N,N-dimethylisopropylamine, and the difference between the localized and delocalized $DMP-L^-$ and $DMP-D^-$ molecules were discussed in detail. The scaled-down SIC idea (i.e., SIC/2) was introduced. Prof. Hannes Jónsson pointed out that CSIC/2 (CSIC = complex SIC) improves atomization energies and bandgaps in comparison to unscaled SIC. In addition, periodic systems like MgO (replacing one Mg with Li) and SiO_2 (replacing one Si with Al) were discussed as success stories where CSIC/2 provides the correct description in contrast to the modern HSE06 functional.

Prof. Susi Lehtola introduced various localization schemes (e.g., Foster-Boys, Pipek-Mizey, Edmiston-Ruedenberg, Fourth Moment, van Niessen, natural bond orbitals) to the audience. Generalized Pipek-Mizey was discussed as an alternative localization method in comparison to Wannier functions for solids. Prof. Lehtola mentioned that PBEsol-CSIC performs better than PBE-CSIC.

Further, the problem of SIC symmetry breaking, i.e., two distinct SIC solutions for each system, was discussed for C_6H_6 and the H_6 molecule.

After the first session, a roundtable discussion offered the possibility for extended questions. The day ended with a poster sessions where young scientists provided detailed insights into their respective SIC research topics. The prize for the best poster was later awarded to the poster "What is right and what is wrong? Spin- and dipole contamination in FLO-SIC" by Sebastian Schwalbe and co-workers (see Fig.2).

Tuesday

The first session of Tuesday focused on recent developments of methods like Koopmans-compliant functionals and applications. Prof. Eric Suraud provided insights into ADSIC and a new two-set SIC and their application to calculating static properties (e.g., ionization potentials) as well as dynamic properties (e.g., photo electron spectra (PES)). The next talk was given by Nicola Colonna, who focused on Koopmans-compliant functionals and their extension with Perdew-Zunger SIC as an efficient tool for the calculation of spectroscopic properties. The current implementation is based on the Car-Parrinello cp.x code as part of the Quantum Espresso program suite. The theoretical foundations (e.g., the IP theorem and piece-wise linearity (PWL)) as well as the application (e.g., GW 100 test set) of this new class of functionals were discussed in detail.

After a coffee break, an alternative approach called "constrained DFT" was introduced by Prof. Stefano Sanvito as a tool for studying charge dynamics in organic materials. It focused on a proper selection of constraints that can provide detailed information on systems with relatively easy calculations. Prof. Nektarios N. Lathiotakis spoke about the constrained search for the optimal local potential in the framework of optimized effective potentials (OEP) offering a way to compensate self-interaction effects. The last talk in this session was given by Dr. Michael Lorke, who presented a new kind of Koopmans-compliant screened exchange potential with correct asymptotic behaviour for semiconductors. His new functional was implemented in a developer version of the VASP code.

An extended lunch break granted the opportunity of taking lunch in old town Freiberg or directly at the conference center. The afternoon session focused on the performance of modern meta-GGA SCAN functional and how SIC-related exact conditions can be used in the construction of hybrid functionals. The session started with Prof. Jianwei Sun, who spoke about self-interaction error and spin symmetry breaking in DFT calculations of d and f- shell complex materials. Prof. Sun mentioned that the intrinsic SCAN self-interaction error is smaller than the PBE self-interaction error. A special emphasis was given to the difference between restricted and unrestricted calculations (e.g., for the H_2 molecule). Further, generalized Kohn-Sham and the question whether spin symmetry breaking is real or artificial were discussed. Especially the application of the SCAN meta-GGA functional to strongly correlated systems (e.g., MnO,

(a) Poster discussions.
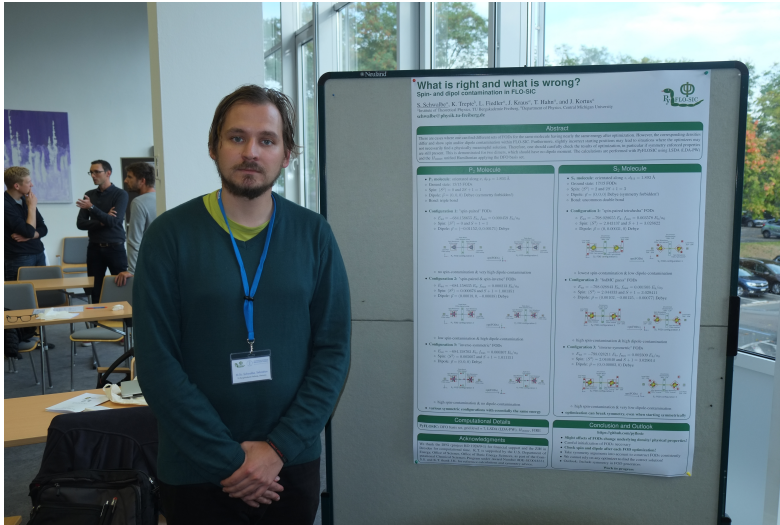
(b) Prize-winning poster.

Figure 2: Poster discussions and prize-winning poster. The prize for the best poster was awarded to Sebastian Schwalbe and co-workers.

FeO, CoO, NiO, and cuprates) were discussed in conjunction with their magnetic ground state. Special quasi-random structures (SQS) for such systems treated with SCAN open the bandgap for paramagnetic phases, but the size of the gap is still underestimated. Prof. Sun noted that SCAN localized the densities around the atoms. The talk of Prof. Sun ended with failures of the SCAN functional, including a too large energy difference between fcc and bcc states for pure Fe.

The next talk was given by Prof. Leeor Kronik. He provided details of SIC-related exact conditions for functionals like no one-electron self-interaction-error, PWL, and correct $1/r$ asymptotic potential for finite objects. He showed how one can include each of these exact conditions in the construction of new hybrid functionals with improved accuracy. The last talk of the day was given by Dr. Kai Trepte, who provided insights into the interpretation and automatic generation of Fermi-orbital descriptors (FODs), which are the key elements in the Fermi-Löwdin orbital self-interaction correction (FLO-SIC) method. Dr. Kai Trepte introduced the chemical interpretation of FODs, with a focus on the so-called FOD bond order.

All conference members attended the conference dinner at the end of this day at the *Ratskeller* in Freiberg, where the international guests could try some German food and beer. Many interesting scientific conversations were carried on until very late that night.

Wednesday

The first part of the morning session on Wednesday added the many-body perspective to the problem, while the second part was focused on applications to solids.

Dr. Pal Daniel Mezei started the day, providing the audience with a many-body perspective on correlation- and self-interaction error within the random phase approximation. He discussed various kernels (e.g., dRPA, ALDA, APBE, NEO, RPAx) as well as direct ring coupled cluster doubles theory (drCCD). The second talk of this day was given by Dr. Giovanni Li Manni, who talked about stochastic CASSCF and MCPDFT for large active space calculations. He introduced active space theories and the limited memory as the bottleneck of these theories. The discussed theories are implemented in OpenMOLE and Molcas. The Full-CI Quantum Monte Carlo (FCIQMC) method was mentioned. Applications of the method to Fe(II) porphyrin and the FeS cluster were discussed. The MC-PDFT theory as a combination of wavefunction theory and DFT was introduced and discussed to solve the question of the ground state of Fe(II) porphyrin.

Dr. Leon Petit introduced a practical approach for the application of PZ-SIC to solids. Full-potential SIC-LSDA and LMTO-ASA and their application to EuO, EuN, NdO, HoSe, SuTe, EuTe, NaCl, and CsCl were discussed in detail. Prof. Thomas Frauenheim, Prof. Gotthard Seifert and Dr. Ben Hourahine gave a detailed introduction to the DFTB method, mentioning applications and successes as well as possible SIC-DFTB implementations.

After the lunch break, Prof. Juan Peralta started a session focusing on the FLO-SIC method. Prof. Peralta introduced FLO-SIC and summarized successes of the method. The next talk was given by one of the inventors of the FLO-SIC method, Prof. Mark. R. Pederson. He started with challenges, simplifications and complexities, ranging in topic from numerics, starting points, diagonalization, variational convergence, Koopmans' theorem, symmetry breaking, the existence of multiple minima, and the meaning of multiple minima to the statements that d- and f- electrons are difficult to describe and finding FODs in metals is a challenge. His talk ended with a discussion about real and complex orbitals. He introduced the density matrix as an intrinsic object. For solving the localization equation there are $N \times N$ unitary transformations, and the action depends explicitly on the density. He explained that FLO-SIC is not an approximation to localization equation (LE)-SIC, and that FLO-SIC is a density matrix theory. Prof. Pederson discussed the possibility of complex-valued FODs. In addition, an outlook was presented for biologically relevant systems like photosystem II, as well as for quantum computing and catalysis. Prof. Rajendra Zope finished the session with a talk about strategies to obtain accurate energies from FLO-SIC. In detail, local and global SIC-scaling variants were discussed.

Thursday

The original timetable was rearranged to concentrate on the hands-on session on Thursday. In total, 9 people joined the practical session. Apart from the young researchers, Prof. Koblar Alan Jackson and Prof. Mark R. Pederson participated as well. The PyFLOSIC code and some auxiliary FOD codes (e.g., fodMC) were tried out by the participants. Additionally, another SIC implementation developed by Aleksei Ivanov (working in the group of Prof. Hannes Jónsson)

- PZ-SIC in GPAW: `https://gitlab.com/gpaw/gpaw/merge_requests/470`,

was discussed and tested too.

Roundtable discussions

The meeting provided several time slots for roundtable discussions. The discussions were very productive with lots of participation and many good ideas were exchanged. We believe that this was one feature of the meeting that made it quite successful. These discussions were especially beneficial to the younger scientists attending the meeting, allowing them to ask specific questions and getting very profound answers from the more senior participants.

The roundtable discussions provide a very good summary of the meeting's scientific presentations.

- Self-interaction error is widely recognized as a problem for standard density functional theory methods. An array of methods that address this error were presented and discussed at the meeting, including PZ-SIC and related approaches.

- PZ-SIC has had many successes for one-electron properties such as orbital energies and band gaps or improved description of anions. Other properties such as atomization energies have been more problematic, but the use of complex orbitals paired with appropriate functionals may provide a way forward.

- Schemes to scale SIC may also provide another way forward, by reducing a tendency of SIC to over-correct the self-interaction error. Such schemes must be employed carefully to avoid compromising the performance for one-electron properties. Schemes utilizing an iso-orbital indicator to scale the correction locally may hold particular promise by preserving the slowly-varying limit of uncorrected density functional approximations.

- The FLO-SIC method is another new direction for implementing SIC. It may allow applications to larger systems than is currently possible with PZ-SIC. Additional work on technical details may lead to improved efficiency and resolve questions about the uniqueness of solutions in FLO-SIC.

Final Conclusion

Judging from the responses of the participants we feel that the workshop was a success. The ideas of self-interaction correction appear to be important for the development of modern functionals and several applications clearly showed that some materials, which where thought to be strongly correlated ones, can be described properly by means of DFT with some additional corrections derived from ideas related to SIC.