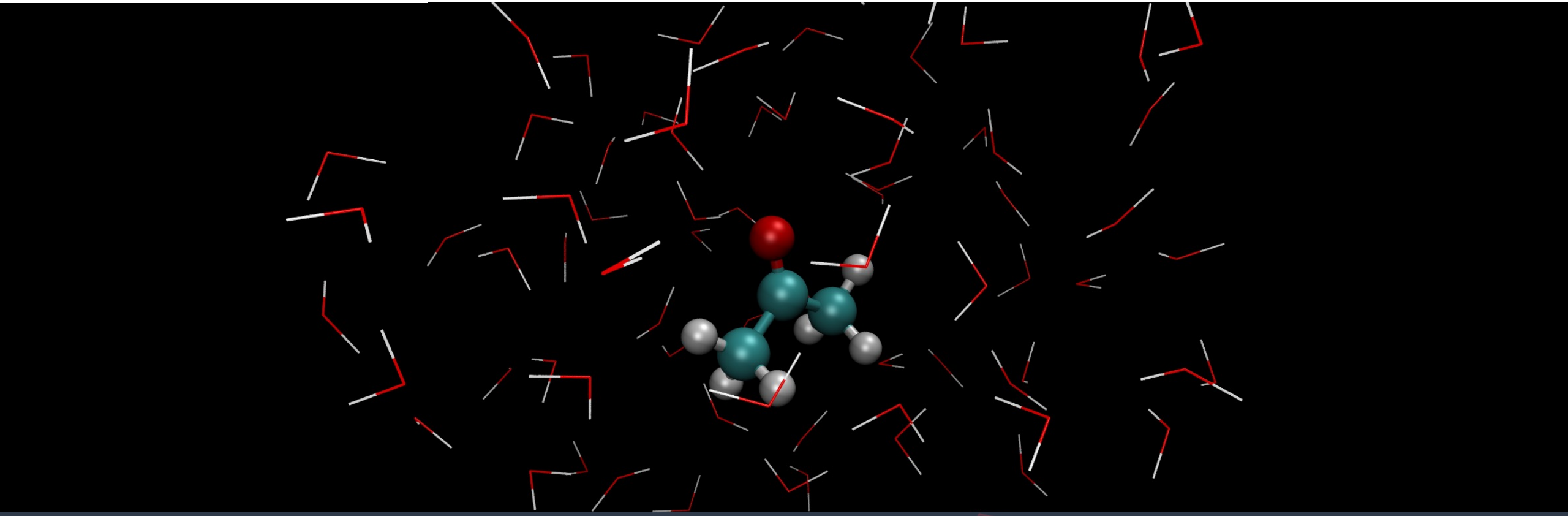
BAUMEIER RESEARCH GROUP

Los Alamos NATIONAL LABORATORY

TU/e EINDHOVEN UNIVERSITY OF TECHNOLOGY



VOTCA and LAMMPS for electronic spectroscopy and transport

LAMMPS Workshop 2021

- Dr. Björn Baumeier / Dr. Christoph Junghans
- Ruben Gerritsen / Vivek Sundaram

votca.org

@VOTCA_software

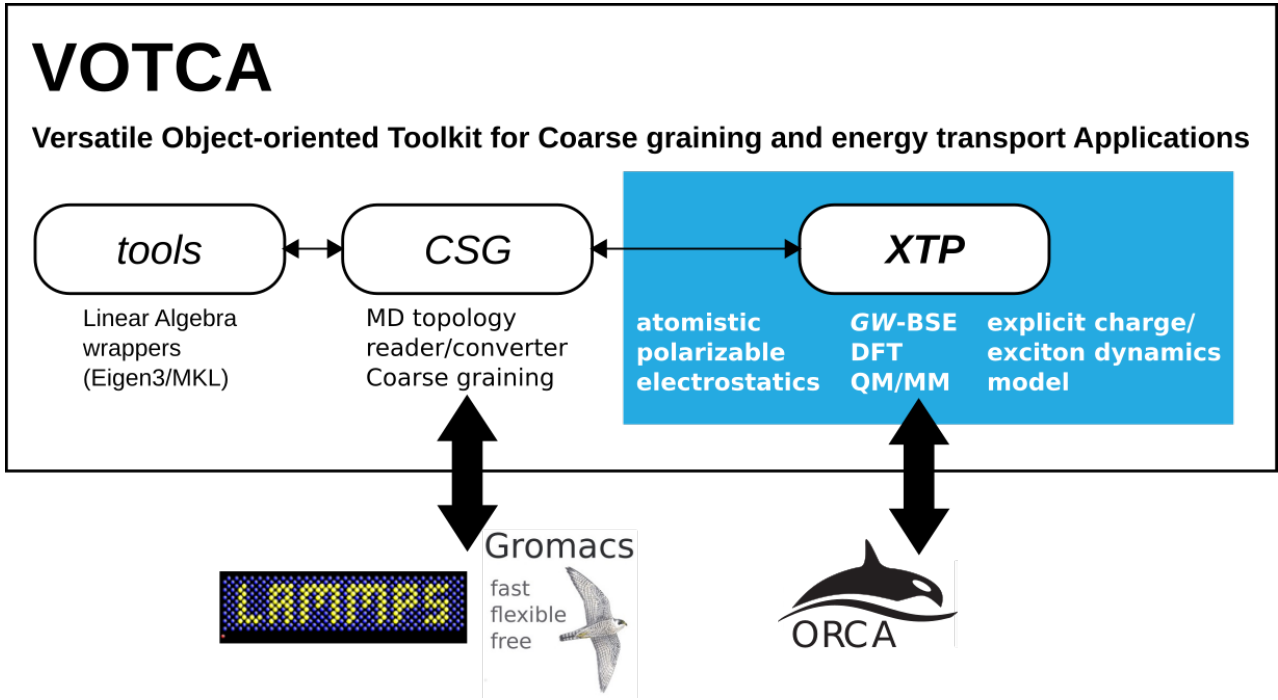
What is VOTCA?

Versatile Object-oriented Toolkit for Coarse-graining Applications
V. Ruehle, C. Junghans, A. Lukyanov, K. Kremer, D. Andrienko
J. Chem. Theo. Comp. **5**, 3211 (2009)

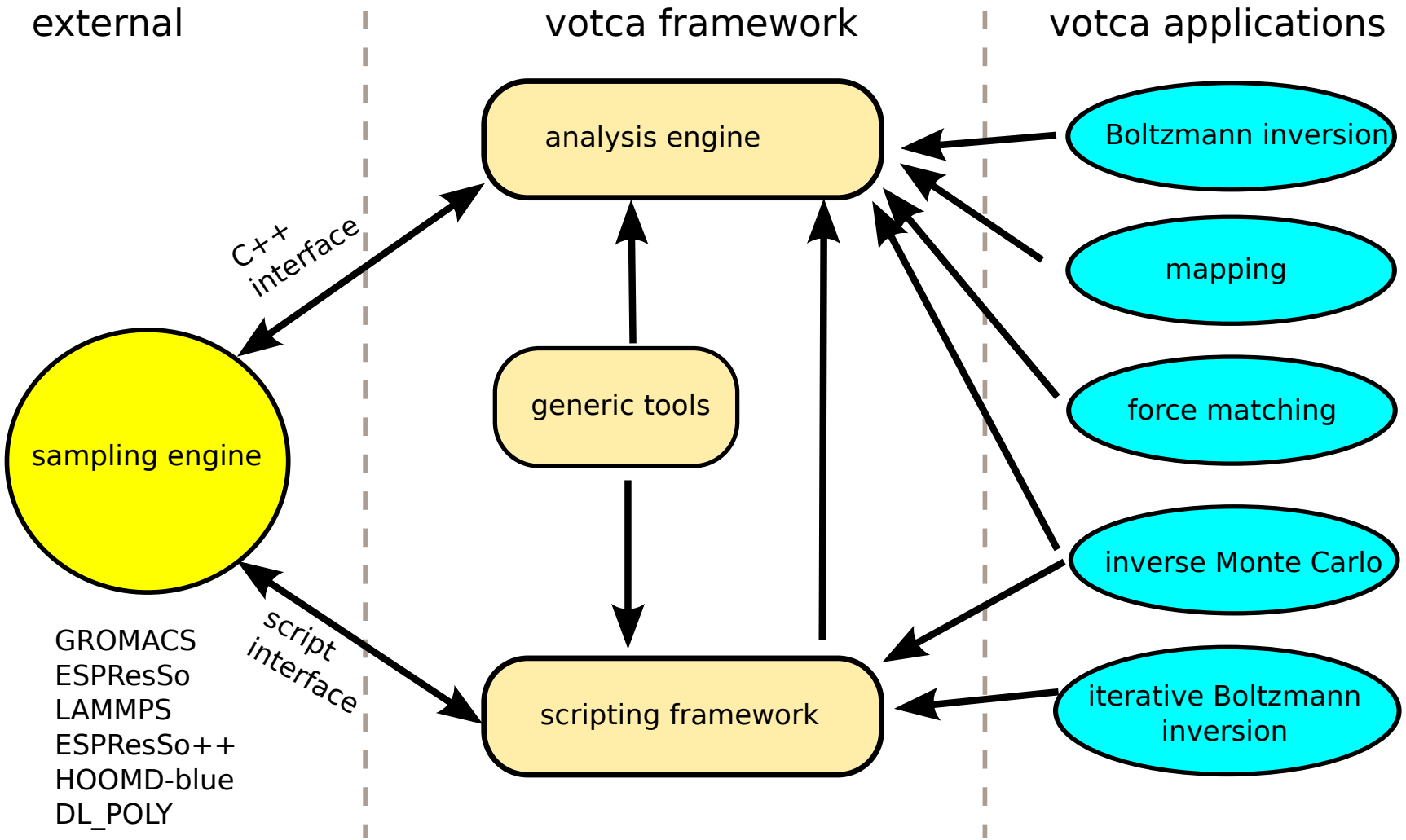
Microscopic simulations of charge transport in disordered organic semiconductors
V. Ruehle, A. Lukyanov, F. May, M. Schrader, T. Vehoff, J. Kirkpatrick, B. Baumeier, D. Andrienko
J. Chem. Theo. Comp. **7**, 3335 (2011)

Electronic Excitations in Complex Molecular Environments: Many-Body Green's Functions Theory in VOTCA-XTP
J. Wehner, L. Brombacher, J. Brown, C. Junghans, O. Caylak, Y. Khalak, P. Madhikar, G. Tirimbo, B. Baumeier
J. Chem. Theory Comput. **14**, 6353 (2018).

Excited-State Electronic Structure of Molecules Using Many-Body Green's Functions: Quasiparticles and Electron-Hole Excitations with VOTCA-XTP
G. Tirimbò, V. Sundaram, O. Çaylak, W. Scharpach, J. Sijen, C. Junghans, J. Brown, F. Zapata Ruiz, N. Renaud, J. Wehner, B. Baumeier
J. Chem. Phys. ***152***, 114103 (2020).



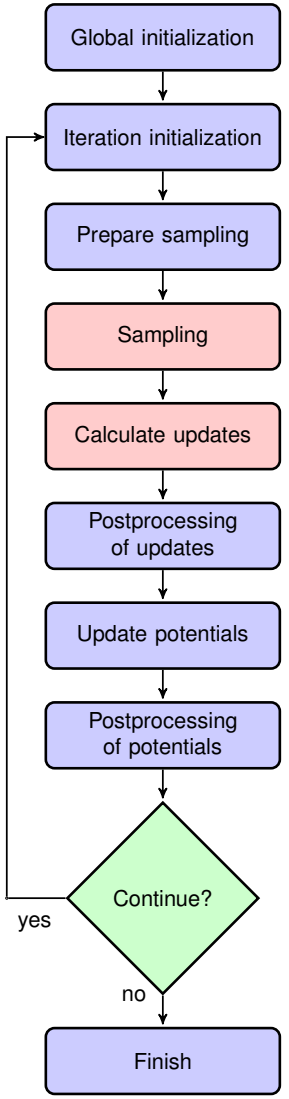
Coarse-Graining in VOTCA



Iterative Workflow in VOTCA-CSG

In short, repeat iterative process until a desired accuracy:

$$U_{n+1}^{CG} = U_n^{CG} + \Delta U\left(n^{th}\ CG\ Simulation\right)$$



VOTCA-CSG offers:

- Steering options in XML file
- Customizable (e.g. replace sampling engine)
- Partially parallelized
- Check-pointed
- Post-processing options:
  - Smoothing
  - Spline fitting
  - Inter/Extrapolation
  - Additional corrections (KB-IBI, C-IBI)

Electronic Transport & Spectroscopy in VOTCA-XTP

Electronic Excitations (Quantum Mechanics)

- Electron detachment (highest occupied molecular orbital, UPS)
- Electron attachment (lowest unoccupied molecular orbital, IPS)
- Electron promotion (electron-hole pair, absorption, emission)
- Electron dynamics (charge hopping, energy transfer)

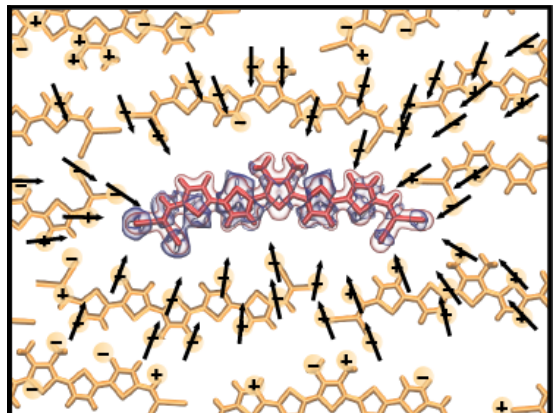
Complex environments (Molecular Mechanics)

- Thin molecular films
- Solute-solvent systems
- Static and dynamic disorder
- …

Many-Body Green's Functions Theory (*GW*-BSE)

- Gaussian-orbital implementation
- Compatible with other GTO packages for DFT ground state
- Hybrid OpenMP/Cuda
- Coupling to external MM potentials

J. Chem. Theory Comput. ***14***, 6353 (2018).



***GW*-BSE/MM**

Polarizable Force Fields

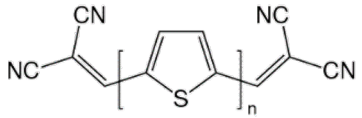
- Distributed atomic multipoles
- Distributed atomic polarizabilities

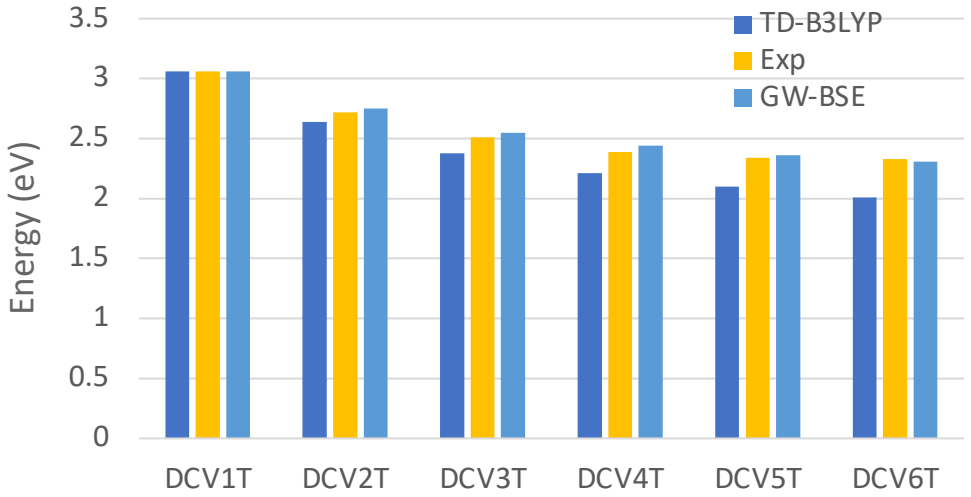
J. Chem. Phys. ***152***, 114103 (2020)

Why *GW*-BSE?

Many formal reasons, but let's just look at some results:

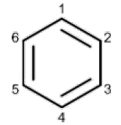
Molecular excitations:



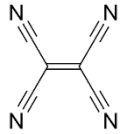


J. Chem. Theory Comput. ***8***, 997 (2012).

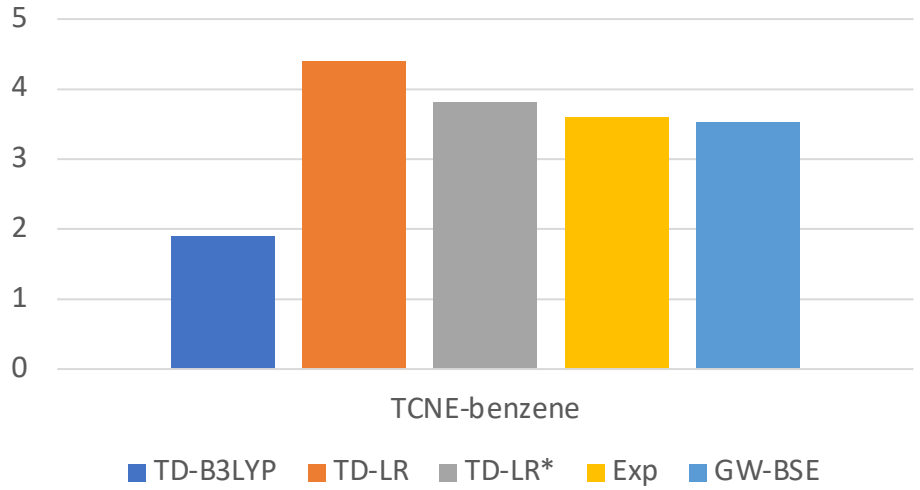
Bi-molecular charge transfer excitations:



Benzene
(donor)



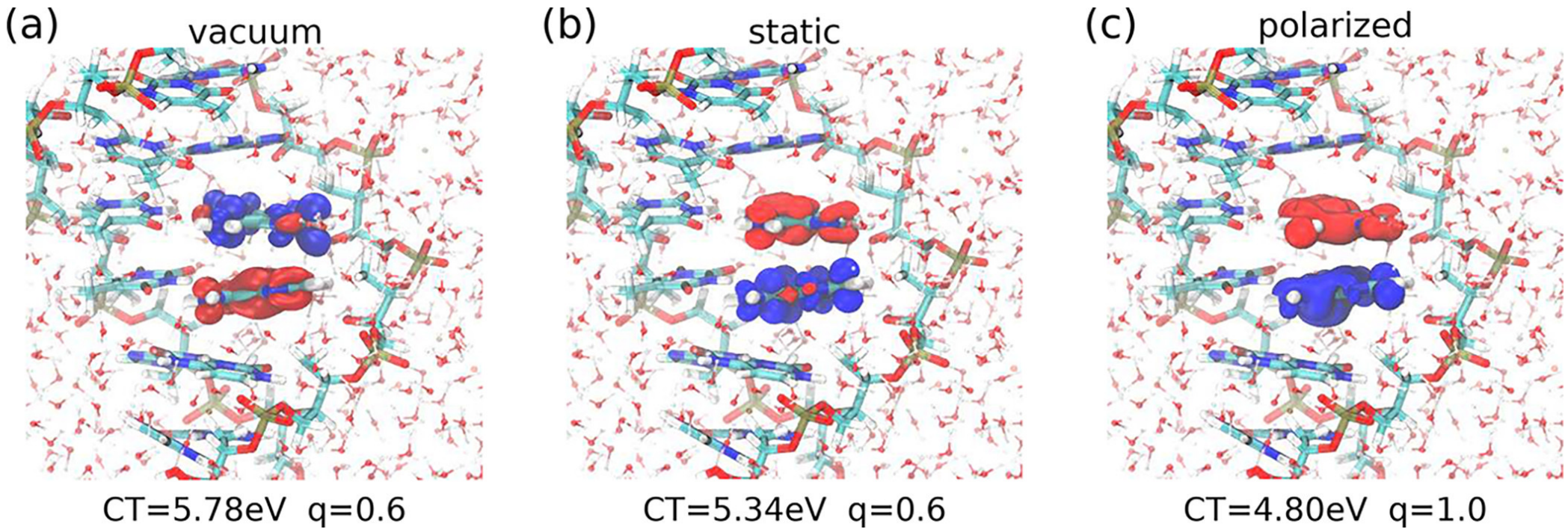
TCNE
(acceptor)



J. Chem. Theory Comput. ***8***, 2790 (2012).

Why a Polarizable Force Field?

Again, an example:

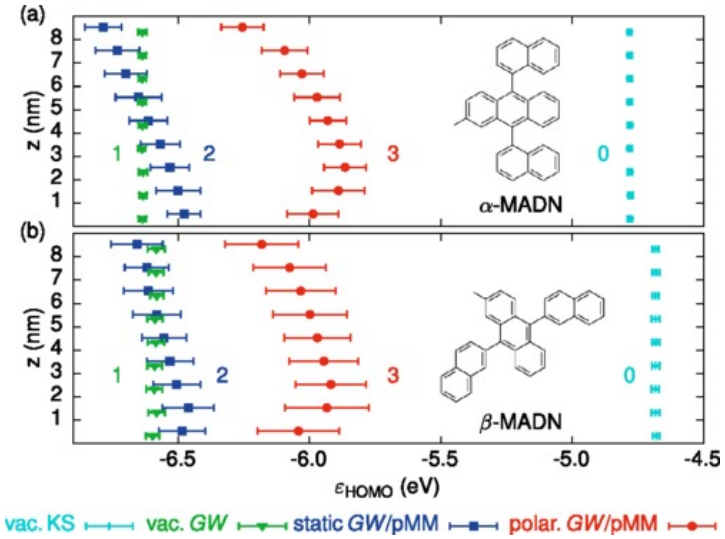


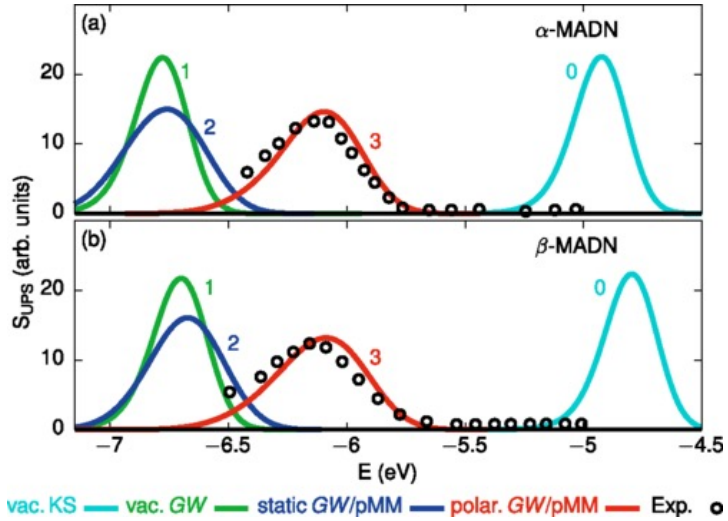
J. Chem. Theory Comput. ***14***, 6353 (2018).

Some More Application Examples

Quantitative predictions of photoelectron spectra in amorphous molecular solids from multiscale quasiparticle embedding

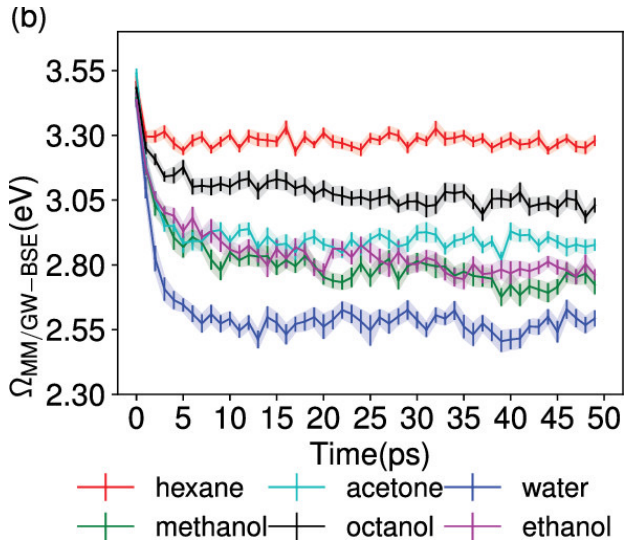
Phys. Rev. B **101**, 035402 (2020)

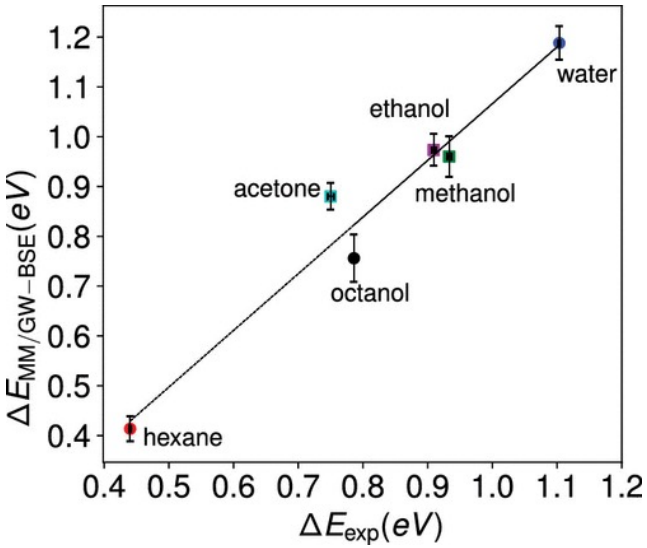




Ultrafast Formation of the Charge Transfer State of Prodan Reveals Unique Aspects of the Chromophore Environment

J. Phys. Chem. B **124**, 2643 (2020)





Check it out on GitHub

votca

http://www.votca.org votca@votca.org Verified

Overview Repositories 30 Packages People 6 Projects

Pinned

votca
Helper to easily install VOTCA-CSG and VOTCA-XTP
CMake 18 15

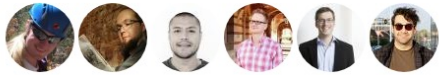
csg
Coarse-graining potentials from atomistic references made easy
C++ 27 25

xtp
GW-BSE for excited state Quantum Chemistry in a Gaussian Orbital basis, electronic spectroscopy with QM/MM, charge and energy dynamics in complex molecular systems
C++ 28 18

csg-tutorials
VOTCA-CSG coarse-graining tutorials
Shell 1 12

xtp-tutorials
VOTCA-XTP spectroscopy and carrier/excitation calculation tutorial
Jupyter Notebook

People



Top languages

Shell Python C++ HTML CMake