

# Accuracy of electronic structure calculations

P.Blaha

Inst. Materials Chemistry, TU Vienna, Austria

The accuracy of electronic structure calculations depends on two factors: First of all on the specific approximation used for the unknown exchange-correlation functional within Kohn-Sham (KS) density functional theory. There are many different approximations like the local density approximation, numerous generalized gradient approximations (GGA), meta-GGAs, hybrid functionals, DFT+U or van der Waals functionals. All of them have their merits and are quite successful for certain applications, but none of them has reached « chemical accuracy » (0.4 eV/atom) for all possible systems or properties. Typically, accurate functionals for solids are less accurate for molecules, functionals yielding good geometry (bond distances) may not give good energetics (cohesive energy) and vice versa. The use of different functionals for different systems/properties leaves the truly « ab initio » way of electronic structure calculations. In Ref. [1] we have compared a large number of different functionals and assessed their accuracy. Recently we extended these studies to various van der Waals functionals [2]. In general, the best functionals yield MARE of about 0.5 % for lattice parameters and 4 % for binding energies for strongly bound systems, while the errors for weakly bound systems, even when using dedicated van der Waals functionals, are 2 and 4 times larger, respectively. Perdew et al. [3] showed recently, that our « standard » PBE-GGA makes a wrong structure prediction for about 20-30% of all investigated systems, while the SCAN meta-GGA improves a lot for main group compounds, but the reliability for transition metal compounds is unfortunately still limited.

The second factor concerns the solution of the KS equations by a particular method implemented in a specific computer code. Roughly speaking, fast but less precise plane-wave pseudopotential methods compete with slower but in general more reliable all-electron codes like WIEN2k [4]. It is worth noting that using a particular code does not guarantee precise results, as all codes have various options to increase precision or, alternatively speed-up the calculations. This ranges from using different pseudopotentials to various series truncations, numerical grids or additional basis functions.

## References

- [1] F. Tran, J. Stelzl, P. Blaha, *J. Chem. Phys.* 144, 204120 (2016).
- [2] F. Tran, L. Kalantari, B. Traore, X. Rocquefelte, P. Blaha, *subm.*
- [3] Y. Zhang et al., *npj Comput. Mater.* 4, 9 (2018)
- [4] Wien2k (<http://www.wien2k.at>)