

Black-box algorithms and robust error control for density-functional theory

Systematic first-principle calculations on thousands to millions of compounds have become an established tool in materials modelling. In this setting automation and reliability of simulations are of elevated importance, which translates to a requirement for numerical methods to be parameter-free and above all robust. At the same time not all calculations in such a screening workflow are required at equal accuracy. A promising outlook of the mathematical study of simulation errors is therefore not only reduce failure rates, but also to identify where approximate computing can be safely employed for gaining extra efficiency in simulations.

Motivated by this prospect this talk focuses on Kohn-Sham density-functional theory (DFT), the most widespread first-principle simulation method in the field. First we discuss the non-linear eigenvalue problem underlying DFT and propose two improvements to the standard self-consistent field (SCF) procedure commonly used for solving it: a preconditioner based on the local density of states as well as a line-search technique based on an approximate model for the DFT energy. Both methods are inspired from a mathematical analysis of the SCF problem, free of user-chosen parameters and robustly applicable to a wide range of systems. With a seamless integration into standard acceleration techniques (such as Anderson) these methods are moreover almost as fast or even faster than standard SCF approaches.

In the second part we discuss error estimation techniques for Kohn-Sham models. For an important quantity of interest, the band structure, we will present guaranteed a posteriori error bounds for the discretisation error in finite plane-wave basis sets. Finally we sketch recent efforts to integrate solid-state DFT simulations with algorithmic differentiation. These techniques enable to automatically compute arbitrary derivatives of output quantities with respect to parameters of the model (DFT functional, pseudopotential etc.), i.e. without needing to implement such derivatives by hand. This is illustrated by exemplary computations tracking the sensitivity of DFT lattice constants on the chosen DFT functional.