## Abstract

**Stochastic Basis Set Approach to Density Functional Theory**

Although density functional theory (DFT) is routinely used to study the properties of a wide range of molecular and crystal systems, its utility for studying large systems in materials science and biology is limited.
This is due to the fact that the DFT computational effort, in terms of memory and CPU time, typically scales cubically with system-size. While linear-scaling methods have been developed by several groups, these are often of limited applicability due to the non-locality of the density matrix in many types of large systems. An alternative approach has been developed which circumvents the calculation of the density matrix and the Kohn-Sham (KS) orbitals using stochastic orbitals. These stochastic orbital methods are useful for orthogonal basis-sets, such as plane-waves and grids. In this work we extend the methods to non-orthogonal basis sets which are common in quantum chemistry. Various observables are treated as random variables where the mean converges to the deterministic KS values as more and more stochastic orbitals are taken. The characteristics of bias and variance with increasing number of random orbitals, as well as with the introduction of density fragments is discussed for observables such as the energy, forces and density of states for water and silicon clusters.

Marcel Fabian

Hebrew University of Jerusalem