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Surrogate Models in Electronic Structure Calculation: Uncertainty Quantification and Propagation

Abstract:
In the last several decades, Density Functional Theory (DFT) has become the most widespread technique to study materials at the atomic scale. Despite its many successes, approximations are required in the implementation, which introduces errors in its predictions. As multi-scale material applications are built, these error from DFT results cannot be ignored and need to be considered as they will propagate to coarser scale models introducing an additional level of uncertainty.
In this work, we have considered one of the main sources of error in DFT modelling, the exchange-correlation energy, and built a surrogate for it using a Bayesian approach which allows for the estimation of the uncertainty in the results from the inability of the considered family of functionals (meta Generalised Gradient Approximation) to reproduce the experimental results. This uncertainty has then been propagated to different relevant material predictions used for different applications. In particular, we have calculated the uncertainty in bulk elastic properties such as lattice constants and bulk moduli and also in the band structure, which can be used in, e. g., electron transport calculations, directly or through coarse grain models using derived parameters such as band-gap and effective mass.
As a second application we have considered propagation of uncertainty to the calculation of alloy thermodynamic properties. Since fully *ab initio* treatment of the alloy properties would be unmanageable, this is done with a two level model. The cluster expansion method is used as a surrogate model for the DFT results. In order to use this, it first has to be trained for the system of interest. This training process introduces an additional level of uncertainty which is captured through the use of a Bayesian regression model. Using this probabilistic cluster expansion, we have calculated uncertainties in thermodynamic properties of alloys, including phase diagrams.