Accelerating *Ab Initio* Molecular Dynamics with On-the-Fly Machine Learning

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Ab initio molecular dynamics is a powerful tool for accurately probing the dynamics of molecules and solids, but it is limited to system sizes on the order of 1000 atoms and time scales on the order of 10 ps. We present a scheme for rapidly training a machine learning model of the interatomic force field that approaches the accuracy of *ab initio* force calculations but can be applied to larger systems over longer time scales. Gaussian Process models are trained on-the-fly, with density-functional theory calculations of the atomic forces performed whenever the model encounters chemical configurations outside of the training set. We demonstrate the flexibility of our approach by testing it on a range of single- and multi-component systems.

I. INTRODUCTION

Key literature:

1. On-the-fly force fields with GPs [1–3]
2. Gaussian Approximation Potentials (GAP) [4–6]
3. The SOAP kernel [7]
4. Other covariant kernels (two- and three-body) [5, 6, 8, 9]
5. Vector-valued GPs and ICM [10]
6. DFT and Quantum Espresso [11, 12]

II. ON-THE-FLY LEARNING

III. COMPUTATIONAL DETAILS

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