Warwick Centre for Predictive Modelling Seminar Series

Molecular Dynamics with on-the-fly Machine Learning of Quantum Mechanical Forces

James Kermode
School of Engineering
University of Warwick

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LIB1, Lower Level, Main Library

Abstract: The need to produce accurate dynamical representations of chemical processes has been ever increasing in recent years. Density functional theory (DFT) provides a well established framework to do this, notably through first principles molecular dynamics (FPMD) simulations. Standard DFT implementations are typically limited to a few hundred atoms, and to the ~10 ps time scale, dramatically restricting the applicability of the approach. Much larger model systems and longer simulations are achievable using force fields, based on judiciously chosen parametrised functional forms fitted on experimental or FP data. However, for many complex chemical situations accurate “reactive” force fields do not yet exist, would require long development times, and would be hard to validate systematically, particularly for use in truly predictive, extrapolative situations.

In this talk I will present a novel molecular dynamics scheme which combines FPMD and machine learning (ML) techniques in a single information-efficient approach. Forces on atoms are either predicted by Bayesian inference or, if necessary, computed with on-the-fly quantum mechanical (QM) calculations and added to a growing machine learning (ML) database. The resulting force field is accurate and transferable, since QM accuracy can be enforced to any desired tolerance and database completeness is never required. The scheme is also efficient since the frequency of QM calls systematically decreases, eventually falling to zero in simple situations where no novel chemical processes or bonding geometries are encountered during the simulation.

The method is expected to be particularly useful for the simulation of processes where complex but recurring chemical steps are encountered, which can be learned, while time-localised occurrences of new chemical bonding geometries cannot be ruled out, so that a fixed force field is not an option. I will motivate and illustrate the approach using examples of challenging materials modelling problems such as crack propagation and crack-impurity interactions.

More info: http://www2.warwick.ac.uk/fac/sci/wcpm/seminars