Adaptive Kinetic Monte Carlo for Long Time Simulations and Global Optimization

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By searching in an unbiased way for possible thermal transitions from a given state, without preconceived notion of final states or transition mechanism, the long time scale evolution of a system can be simulated using the adaptice kinetic Monte Carlo algorithm [1]. Systematic coarse graining of the free energy landscape is important to avoid being slowed down by fast processes. Within the harmonic approximation to transition state theory (HTST), the challenging task is to find all relevant saddle points on the energy rim surrounding an initial state energy minimum [2]. More generally, within full TST, a high dimensional dividing surface needs to be constructed and optimized using Keck's variational principle. In either case, the exact dynamics can, in principle, be obtained from short time trajectories started at the transition state [3]. This approach has been implemented to large extent in the distributed computing software EON (http://theochem.org/EON) [4] and applications to various atomic and spin systems will be described. A modification of the algorithm can be use for global optimization of objective functions of continuous variables [5]. Application to geothermal reservoir modeling will be discussed.

[1] 'Long time scale kinetic Monte Carlo simulations without lattice approximation and predefined event table', G. Henkelman and H. Jónsson, J. Chem. Phys., Vol. 115, p. 9657 (2001).
[2] 'Efficient Sampling of Saddle Points with the Minimum-Mode Following Method', A. Pedersen, S.F. Hafstein and H. Jónsson, SIAM Journal of Scientific Computing 33, 633 (2011).
[3] 'κ-dynamics: An exact method for accelerating rare event classical molecular dynamics', C-Y. Lu, D.E. Makarov and G. Henkelman, J. Chem. Phys. 133, 201101 (2010).
[4] 'Distributed Implementation of the Adaptive Kinetic Monte Carlo Method', A. Pedersen and H. Jónsson, Mathematics and Computers in Simulation 80, 1487 (2010).
[5] 'Simulated Annealing with Coarse Graining and Distributed Computing', A. Pedersen, J-C. Berthet and H. Jónsson, Lecture Notes in Computer Science 7134, 34 (2012).