Accurate and Computationally Efficient Models for Surface Reactions

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Modelling the kinetics of surface catalysed reactions is essential in many complex chemical processes. However, the majority of microkinetic models employ mean-field approximations, which lead to an approximate description of catalytic kinetics by assuming randomly distributed and spatially uncorrelated adsorbates. On the other hand, kinetic Monte Carlo (KMC) methods provide a discrete-space continuous-time stochastic formulation that enables an accurate treatment of spatial correlations in the adlayer, but at a significant computation cost. In this presentation, we show that the so-called cluster mean-field approach can be used to develop higher order approximations that systematically increase the accuracy of kinetic models for surface reactions by treating spatial correlations at a progressively higher level of detail. We focus on a reduced model for NO oxidation and construct a sequence of approximations of increasingly higher accuracy. By comparing the turnover frequencies of these models with those obtained from KMC simulation, we show that such approximations, while more computationally intense than the traditional mean- field treatment, still achive tremendous computational savings compared to KMC simulations, thereby paving the way for employing them in multiscale modeling frameworks.