

Bond-selective chemical reactivity from first principles

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D202 Seminar room, School of Engineering, 2nd Floor

Abstract: Bond selective chemistry is of great importance for many technological applications, including heterogeneous catalysis, surface and nanostructure functionalization. For instance, achieving high selectivity is one of the major goals in catalysis because this allows for producing preferentially the desired products. Recently, initial-state-resolved measurements using supersonic molecular beams have provided unambiguous evidence for vibrational-mode- and bond-specific reactivity of CH₄ and its deuterated isotopologues on both Ni(111) and Pt(111) surfaces [1-2].

Molecular dynamics (MD) is a powerful tool to investigate reaction dynamics at surfaces. However, an accurate and computationally affordable method for describing surface reaction dynamics of polyatomic molecules including surface atom degrees of freedom (DOF), has been long-time expected.

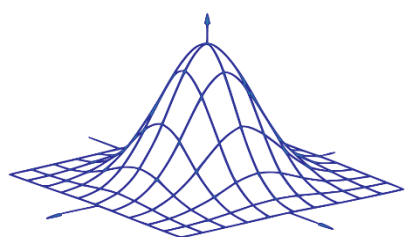
In this talk, I will present results of a quasiclassical MD study of the dissociative adsorption of CH₄ isotopologues on Pt(111), based on an accurate system-specific reactive force field (RFF) parametrized from DFT total energy data [3]. Through simulations treating all the molecular and surface DOFs on an equal footing, we have obtained C-H bond selectivities and vibration-mode-specific reactivities in excellent agreement with experiments.

[1] D. R. Killelea, V. L. Campbell, N. S. Shuman, and A. L. Utz, *Science* 319, 790 (2008).

[2] L. Chen, H. Ueta, R. Bisson, and R. D. Beck, *Faraday Discuss.* 157, 285 (2012).

[3] X. J. Shen, A. Lozano, W. Dong, H. F. Busnengo, and X. H. Yan, *Phys. Rev. Lett.* 112, 046101 (2014).

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