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Semi-empirical approach to the simulation of molecule-surface reaction dynamics

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Propositions

- i. It is not easy to predict and to intuitively explain the behavior of a density functional.
– Chapter 3
- ii. Barrier heights cannot be measured directly, and are best determined through a close comparison of molecular beam experiments and dynamics calculations reproducing the reaction probability measured therein.
– Chapter 4
- iii. The reasons why the two functionals (PBE and SRP32-vdW) predict such different reactivity for CHD₃ on Pt(111) are related to the presence of van der Waals correlation in the SRP32-vdW functional, which is absent from the PBE functional.
– Chapter 6
- iv. The HOD molecule can react regardless of its initial orientation and large changes in the values of the angle θ are observed on the molecule's way to the surface.
– Chapter 7
- v. In principle the SRP-DFT-AIMD approach should be applicable to any molecule reacting with a metal surface, as long as experiments can be devised on the system that are amenable to an accurate quasiclassical dynamical treatment.
– Nattino *et al.*, *J. Phys. Chem. Lett.*, 7, 2402 (2016)
- vi. The CHD₃ molecule does not react by relaxing the reactive bond along the angle θ , but rather by changing the orientation of the umbrella axis after passing through a reactive gate.
– Füchsel *et al.*, *Phys. Chem. Chem. Phys.*, 18, 8174, (2016)
- vii. The AIMD method allows the calculation of statistically accurate dissociation probabilities without the need of introducing *a priori* dynamical approximations concerning the evolution of specific molecular degrees of freedom or concerning the role played by surface atom motion.
– Nattino *et al.*, *J. Chem. Phys.*, 144, 044702 (2016)
- viii. The dissociation of small polyatomic molecules on transition metal surfaces often represents the rate limiting step in the heterogeneously catalyzed processes used to produce chemicals on an industrial scale. Developing a predictive understanding of these dissociation reactions is not only of fundamental interest but also of practical importance.
– Chadwick *et al.*, *J. Chem. Phys.*, 148, 014701 (2018)
- ix. Answers are always limited, temporary, unsatisfactory. Questions, on the other hand, are the true engine of mental activity: a person who does not ask questions, or who is satisfied with the answers, cannot go very far.
“*Le risposte sono sempre limitate, provvisorie, insoddisfacenti. Le domande invece sono il vero motore dell'attività mentale: un uomo che non si pone domande, o che si contenta delle risposte, non va molto lontano.*”
– Piero Angela, Da zero a tre anni, Mondadori, (2017)