Machine-learning-enabled modelling of ultrafast dynamics at surfaces

Reinhard J. Maurer

Department of Chemistry and Department of Physics, University of Warwick, UK

Electronic excitations at surfaces can be created either by electromagnetic radiation or the ultrafast motion of molecules, for example in photocatalysis or hyperthermal molecular scattering. Such excitations, while short-lived, will trigger coupled and concerted nonadiabatic motion of electrons and nuclei at short time scales that can measurably affect chemical dynamics by introducing energy dissipation, dynamical steering effects, and by contributing to state-dependent reaction probabilities. [1]

I will present our recent efforts to establish methods able to capture nonadiabatic and excited-state effects during ultrafast dynamics at surfaces. We employ a range of methods such as molecular dynamics with electronic friction and surface hopping dynamics. [2] By combining first principles electronic structure calculations [3] with high-dimensional machine-learning representations, [4,5] we are able to simulate measurable observables with statistical averaging over thousands of reaction events. Doing so enables us to directly compare our results to experiments and to assess the limitations of different methods. Recent relevant results include the vibrational state-to-state scattering of H_2 on copper surfaces [5], NO on Au(111), [6] and the light-driven hydrogen evolution reaction.

[1] Bartels et al., *Chem. Sci.*, **2011**, *2*, 1647−1655
[2] Gardner et al., *J. Chem. Phys.* **2022**, *156*, 17480, https://github.com/NQCD/NQCDynamics.jl
[3] Box et al., *IOP Electronic Structure*, **2023**, *5*, 035005
[4] Stark et al., *Mach. Learn.: Sci. Technol.*, **2024**, *5* 030501
[5] Sachs et al., *Mach. Learn.: Sci. Technol.*, **2025**, *6*, 015016
[6] Meng et al., *Phys. Rev. Lett.*, **2024,** *133*, 036203