Accessing photoinduced reaction dynamics on surfaces with neural networks

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Modelling the ultrafast photo-induced dynamics and reactivity of adsorbates on metals requires including the effect of the laser-excited electrons and, in many cases, also the effect of the highly excited surface lattice. Ab initio molecular dynamics with electronic friction and thermostats, (Te,Ti)-AIMDEF do enable such a complex modelling [1], but, unfortunately, these simulations come with a very large computational expense that severely limits statistics and propagation times.

In the last years, the use of neural network (NN)-generated multidimensional potential energy surfaces (PESs) and, in particular, the use of the atomistic neural network (AtNN) approach [2], is becoming the accurate alternative to ab initio molecular dynamics studies of diverse gas-surface reactions, including the dynamics at the solid-liquid water interfaces. However, it must be emphasized that the requirements imposed to a NN-PES capable of describing photo-induced reactions are extremely demanding as compared to those required in usual elementary gas-surface processes.

A reliable NN-PES must be able to model large movements of multiple and different adsorbates and surface atoms and it must describe accurately the very distinct and changing adsorbate coverages that exist during the photoinduced dynamics. This means that it is necessary to assure a precise description of adsorbate-substrate and interadsorbate interactions under very different and changing conditions, including local variations in the number of neighbor adsorbates and strong lattice distortions, since the lattice temperature may vary rapidly in the range 90-1000 K. Therefore, altogether the requirements for the atomistic NN-PES are unprecedentedly extreme and demanding for this kind of processes.

In this talk I will show good examples of accurate NN-PESs that we have constructed using different atNN approaches [3], including the recently developed embedded atom neural network (EANN) method [4], which we find to be impressively accurate and flexible to account for all the necessities required to deal with photoinduced reactions [5]. Neural networks allowed us to understand the strong coverage dependence found experimentally in CO/Pd(111) [6], the large branching ratio between CO desorption and CO oxidation in Ru(0001) [7], and reveal the dynamical nature of the CO physisorption well that so far was only found in XPS experiments.

References

[1] M. Alducin, N. Camillone, S.-Y. Hong, J. I. Juaristi, Phys. Rev. Lett., 123 (2019) 246802.
[2] J. Behler and M. Parrinello, Phys. Rev. Lett., 98 (2007) 146401.
[3] A. Rivero Santamaría, M. Ramos, M. Alducin, H.F. Busnengo, R. Díez-Muiño, and J.I. Juaristi, J. Phys. Chem. A, 125 (2021) 2588.
[4] Y. Zhang, C. Hu, B. Jiang, J. Phys. Chem. Lett. Journal, 10 (2019) 4962.
[5] A. Serrano Jiménez, A. S. Muzas, Y. Zhang, J. Ovčar, B. Jiang, I. Lončarić, J. I. Juaristi, and M. Alducin, J. Chem. Theory Comput. 17 (2021) 4648.
[6] A. S. Muzas, A. Serrano Jiménez, Y. Zhang, B. Jiang, J. I. Juaristi, and M. Alducin, J. Phys. Chem. Lett. 15 (2024) 2587.
[7] I. Žugec, A. Tetenoire, A. S. Muzas, Y. Zhang, B. Jiang, M. Alducin, J. I. Juaristi, J. Amer. Chem. Soc. Au, 4 (2024) 1997.

Figures

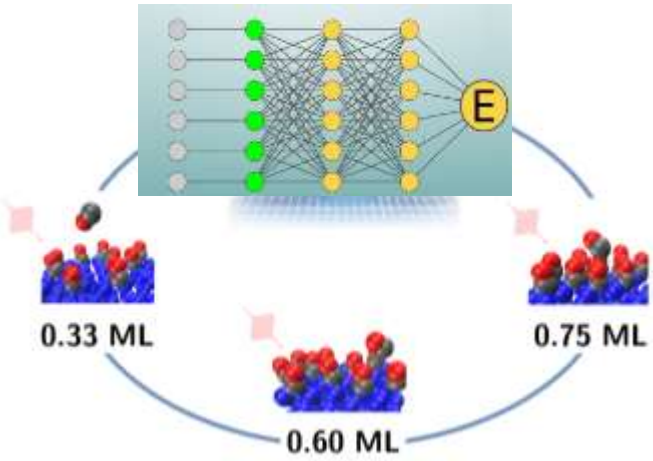

Figure 1. Scheme of a Neural Network Potential Energy Surface valid to describe photoinduced reactions on different covered surfaces.

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