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Electron-Hole Pairs during Adsorption Dynamics of O_2 on Pd(100) – Exciting or not? JOERG MEYER, KARSTEN REUTER, TU Muenchen — *Ab initio* modeling can provide important atomistic insights into the dynamics of elementary reaction steps in heterogeneous catalysis. But already an apparently simple example like the adsorption of O_2 on a frozen Pd(100) surface brings up several fundamental challenges: The corresponding six-dimensional potential energy surface (PES) is required, and excitations of electron-hole (eh) pairs as well as the transition from the $^3\Sigma_g^-$ spin-triplet in the gas phase to a singlet-like state of adsorbed oxygen might require to go beyond the Born-Oppenheimer approximation. We tackle the PES by neural network interpolation, based on a coordinate transformation that correctly includes and exploits the underlying symmetry. Classical molecular dynamics thereon yields the initial sticking coefficient in good agreement with experimental data. Scrutinizing this result, we calculate spin-resolved eh pair spectra for several trajectories of different statistical relevance using a computationally appealing perturbative approach building on time-dependent DFT.[1] Although non-adiabatic energy losses do not exceed 5% of the chemisorption energy, their importance for the spin transition is finally discussed.

[1]J. Meyer and K. Reuter, New J. Phys. **13**, 085010 (2011)

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