Water and hydroxides at interfaces – similar and different

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Intact and dissociated water, i.e. H_2O and OH^-, on metal oxide and mineral surfaces are found in a far larger variety of –often strained– structural motifs than what is observed in bulk hydrates and hydroxides crystals. For the bulk crystals, robust structure–vibrational frequency relations have been established in the literature, but for surface systems, almost none exist. Using theoretical calculations, we have examined surface-adsorbed H_2O and OH^- groups to search for descriptors (features) that manage to express – or predict – the vibrational frequency of the OH surface species; cf. **Fig. 1**. Intact and dissociated water molecules can demonstate quite different correlation curves, which can in fact be an aid when correlation curves are used to decipher experimental spectra.

Furthermore, starting from a machine-learning geometrical descriptor, we have examined how the frequency prediction power (and "insight content") vary when more physics/chemistry flavor is gradually included (**Fig. 2**).

I will also discuss some results from thick water films ("solid-liquid interfaces") such as why ceria with a lot of water "on top" can be hydrophobic while the direct ceria-water interaction is in fact hydrophilic [3].

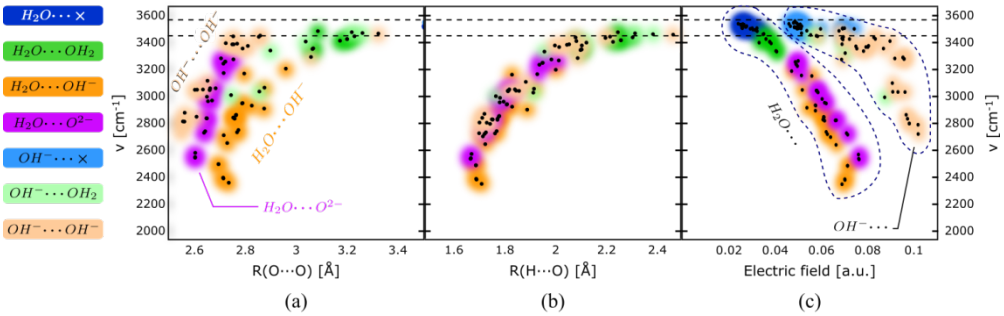

Fig. 1. 'OH frequency-structure' correlations at a water-ceria(111) interface from periodic DFT calculations folllowed by quantum-vibrational calculations (cf. Ref. [1]). Coverages from 0.25 ML up to ≈2 ML are included.

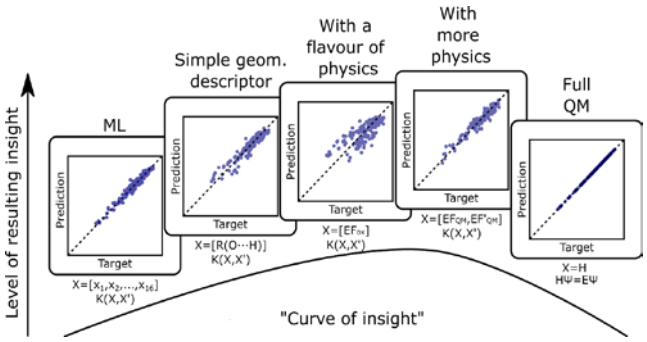

Fig. 2. Progression of our descriptors in terms of the "amount" of physics coded into them and their respective level of insight. The scatter plots show their capability of predicting vibrational frequencies (cf. Ref. [2])

[1] A. Röckert, J. Kullgren, K. Hermansson, J. Chem. Theory Comput, **18**, 7683 (2022).
[2] J. Kullgren, A. Röckert, K. Hermansson, J Phys. Chem. C, Accepted 12 April (2023).
[3] L Agosta, D Arismendi, M.Dzugutov, K Hermansson, Angewandte Chemie Int. Ed. (Comm) 3 April (2023).