Ab initio description of oxides in electrochemical environment by constructing defect and surface phase diagrams

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Electrochemistry offers a huge number of possibility to tune materials properties desired in the context of practical problems such as water electrolysis and batteries, electro-catalysis, photo-catalytic water splitting or corrosion. The quantitative description of materials behaviour presents, however, challenges because of the involvement of different length scales, time scales and materials classes. Our recently developed unifying approach for semiconductor defect chemistry and electrochemistry [1] naturally links ab initio calculations to experimental observables such as pH and electrode potential U, enabling us to characterise materials properties in electrochemical environment. One way in which we can utilize the approach is the construction of relevant Pourbaix-type diagrams, which enable us to discuss the stability of an oxide and its surface reconstructions in electrochemical environment, i.e. against pH and U. Two examples will be presented. Defect Pourbaix diagrams revealing the dominant point defects governing the growth and dissolution of the oxide barrier layer forming when Zn comes into contact with a corrosive environment [2]. Surface Pourbaix diagrams showing stable surface reconstructions forming on the polar Zn-terminated ZnO(0001) surface and which reveal the that solvation effects are substantial, highly selective and play an active role in shaping the surface [3, 4].

[1] M. Todorova and J. Neugebauer, *Phys. Rev. Applied.* **1**, 014001 (2014).
[2] M. Todorova and J. Neugebauer, *Faraday Discuss.* **180**, 97 (2015).
[3] S. Yoo, M. Todorova und J. Neugebauer, *Phys. Rev. Lett.* **120**, 066101 (2018).
[4] S. Surendralal, M. Todorova, M. Finnis und J. Neugebauer, *Phys. Rev. Lett.* **120**, 246801 (2018).