Final Report
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Title: Investigations of charged interfaces for electric vehicle applications

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Abstract:

We refined the parametrization of our model for the hydrogen evolution on Pt(111) in the presence of SO4H2 [1,2,3,4]. In particular we studied the effects of changing the turning point of the water. The form of the voltammogram is not very sensitive to the exact position of the turning point, which in previous work was treated as an adjustable parameter. Physically a reasonable choice is that the turning point is the PZC (point of zero charge). This new parametrization clarified some issues related to the mechanism of hydrogen oxidation. In the early stages of this project, we tried with N. Marzari [5] to compute the ex-situ structure of the bisulfate-water $\sqrt{3}\otimes\sqrt{7}$ (or also $5/2 \otimes\sqrt{7}$) phase, seen by STM. However, we were unsuccessful in reproducing the known structures of the phase, even though we used state of the art pseudo potentials. There are various possible reasons why this happens, but the most obvious one is the environment of the electrode surface. This means that we need a theory that is able to include the local microfield as a function of the applied potential and electrochemical environment, and hence, we need to fully develop the density functional of a real molecular solvent solution in the double layer. A classical functional density theory capable of treating water and ionic solutions near charged electrodes and pores was developed using

Classical Density Functionals. The general theory of Yukawa fluids was useful in formulating such a density functional. A step in this direction is the new scaling mean spherical approximation for the ion-dipole mixture [6]. We have also reformulated the solution of the multi-Yukawa closure of the Ornstein-Zernike equation [7, 8, 9], and in particular found a new solution of the one component Yukawa fluid [10], which satisfies correct symmetry requirements. We have solved the multi-component, multi-density Yukawa mixture [11].

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