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In situ surface X-ray diffraction studies of the electrochemical double layer on Pt(111)

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The molecular structure of the electrochemical double layer is an important fundamental topic in interfacial electrochemistry. It has received renewed interest in the last years because of its influence on many electrochemical reactions and the emergence of ab initio molecular dynamics (AIMD) simulations that allow a detailed description of the interface structure and dynamics [1,2]. In the measurements presented here, we use high energy surface X-ray diffraction (HESXRD) to investigate the properties of the electrochemical double layer on single-crystal Platinum electrodes.

Platinum is widely used in electrocatalytic applications due to its high ORR reactivity, for example in PEM fuel cells. The Pt(111) surface is especially interesting for fundamental studies of the electrochemical double layer. Recent electrochemical measurements revisited this system and found evidence of a double layer structure that strongly deviated from the traditional Gouy-Chapman-Stern model. A structural influence of interfacial water has been proposed as a possible explanation for the deviations close to the potential of zero charge (PZC) [3,4]. Subsequent AIMD simulations indicate a voltage-dependent formation of layers of interfacial water molecules at distinct Pt(111)-water distances between 2-3 Å [5].

To investigate the arrangement and orientation of the water molecules near the Pt(111) surface we used in situ HESXRD methods, focussing on X-ray reflectivity (XRR) and crystal truncation rod (CTR) analysis. These methods allow to acquire detailed information about the electrode surface structure, as demonstrated for studies of the Pt oxide structure [6]. By measuring multiple CTR sets at different surface potentials, information on the potential-dependence of the interface processes can be obtained. The experiments were performed at beamline ID31 of the European Synchrotron Radiation Facility using photon energies of 75 keV. The fast, high-quality data acquisition allows significantly improved structural modeling compared to conventional SXRD. We will report first results on the structure of the interfacial water in the double layer obtained in perchloric acid at potentials around the PZC.

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Interfaces during chemical reactions and catalysis

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