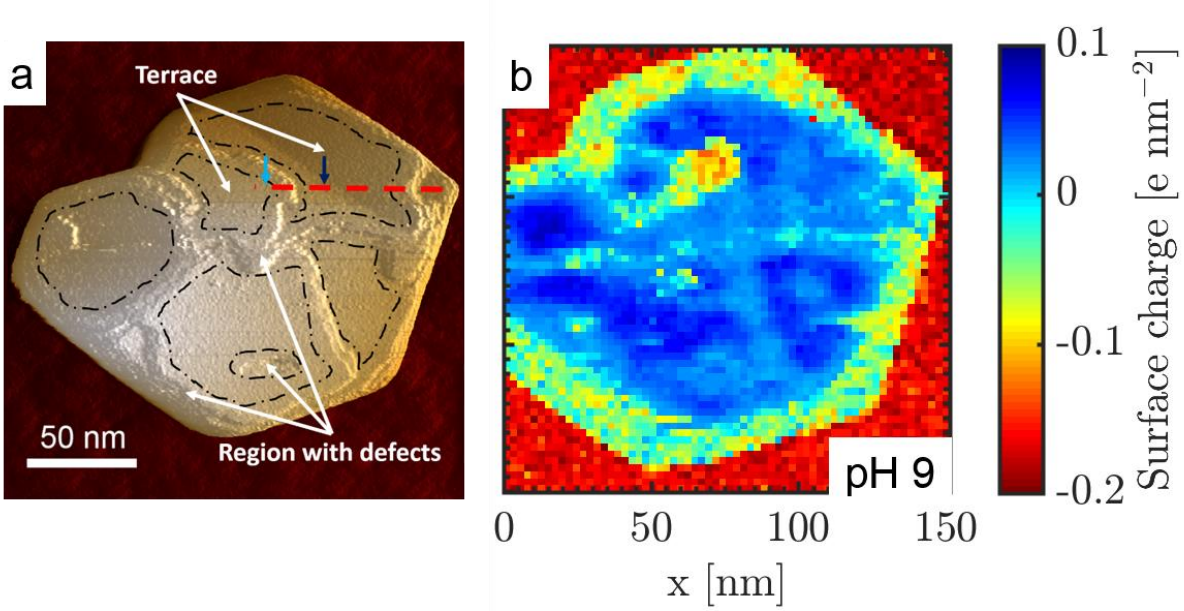
Gibbsite minerals are important for many natural phenomena and technical processes, where they play a crucial role for the retention of nuclear waste and the recovery of oil from geological reservoirs. Their role in these applications primarily originates from their ability to bind organic and inorganic solutes from their surroundings, which is governed by surface charge. Therefore, extensive efforts have been devoted to the characterization of the charging behavior of gibbsite. Despite these efforts, the origin and amount of surface charge on gibbsite is still not well established. In this work we try to identify whether the basal plane of gibbsite does carry charge and if so, determine whether this charge is pH-responsive and to what extent it can be related to the presence of defects on the surface. To this end, we performed dynamic AFM spectroscopy measurements in various aqueous electrolyte solutions on individual gibbsite nanoparticles. The local surface charge is extracted from force-distance measurements using DLVO theory including charge regulation. Our measurements reveal the highly anisotropic and pH dependent surface charge properties of the basal plane of gibbsite. The effective surface charge of defect sites display a stronger pH dependence comparing to adjacent apparently atomically smooth regions of the basal plane. Regions with high defect density seem to reverse their charge within the range of pH 6 to 9, and display a negative charge of -0.10 e/nm2 at pH 9. In contrast, the smooth regions display a positive charge of 0.05 e/nm2 that hardly changes with pH. These measurements demonstrate the role of surface defects for the average surface charge of the gibbsite basal plane.




a) AM-AFM topography image of a gibbsite particle on silica reveals the presence of smooth terraces and regions with defects on the gibbsite basal plane. b) The surface charge distribution of the same particle is homogeneous for silica (red) and heterogeneous for the gibbsite particle (blue and yellow). The regions of reduced charge correlate with the terrace steps and defects in a).