DFT simulations of combined 3D NC-AFM and STM imaging of the Cu(100)-O oxide surface

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In a bid towards functional imaging, three-dimensional atomic force microscopy (3D-AFM) in non-contact mode [1] has been combined with scanning tunnelling microscopy (STM) to study the oxygen-terminated copper (100) surface. Complex 3D data sets, obtained by simultaneously recording the tunnelling current and the AFM frequency shift, contain a remarkable wealth of information. Still, the interpretation of the wide range of contrast modes requires a thorough characterisation of the sources of contrast in AFM and STM imaging.

We combined density-functional theory (DFT) calculations with Non-Equilibrium Green's function (NEGF) methods for electronic transport to determine the tip-surface interaction and tunnelling current [2, 3, 4] for a large set of tip models in order to clarify the different contrast modes obtained in the experiments. We studied the features of a stable Cu(100)(2√2×√2)R45°-O surface reconstruction and identified prospective reaction sites, before introducing model AFM tips to conduct a series of tip approach simulations. The effect of tip changes on imaging modes was explored by considering tips of different reactivity. Our simulations, when related to a particular AFM imaging mode (fig. 1b), revealed that the experimental tip had been Cu-terminated (fig. 1a). Charge density and current calculations further helped us investigate the STM imaging modes for tips of different reactivity. We were able to confirm independently the Cu termination of the experimental tip (fig. 1d) in the STM measurement concurrent with the AFM result (fig. 1c), but also explain a variety of unusual STM contrasts observed. Lastly, surface defect features in the STM image that were absent in the AFM channel led us to consider different point defect models for the Cu(100)-O surface. The defect was identified as the lateral rearrangement of Cu atoms on the surface, a deformation that can further be related to the nucleation and growth of surface domains. The combination of conductance calculations with total energy methods provides insight into the fundamentals of multi-channel SPM imaging, as well as the correlation between measurement and function, making it key to the development and application of this experimental technique to characterise the electrical and catalytic response of new materials.

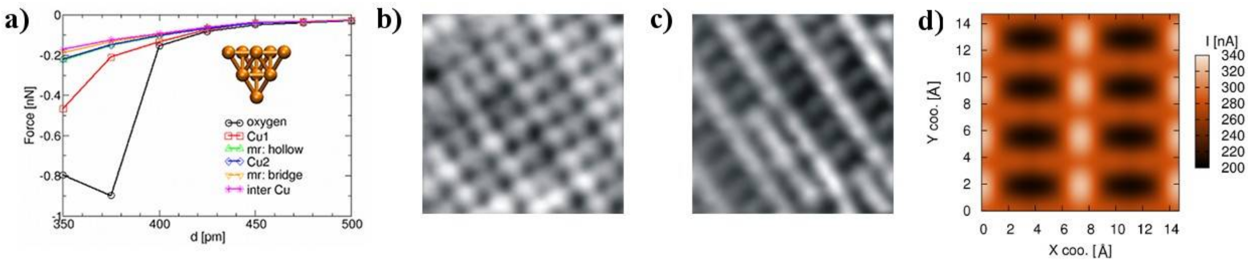

Figure 1: Concurrent AFM and STM experimental images with equivalent theoretical results

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