

METAL ATOMS ON ULTRATHIN INSULATING FILMS: CONTROLLING THE CHARGE STATE OF INDIVIDUAL GOLD ATOMS

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Ultrathin insulating films on metal substrates are unique systems to use the scanning tunneling microscope to study the electronic properties of single atoms and molecules, which are electronically decoupled from the metallic substrate. Moreover the high lateral resolution in combination with local electronic spectroscopy allows also the investigation of the underlying metal/insulator interface. Compared to bulk insulators few atomic layer thick insulators prevent surface charging, strain and image charge effects can be tailored by a suitable selection of the layer thickness and the metal substrate combination, and if applicable the scattering of interface electrons can be exploited to characterize adsorbate insulator interaction.

We have investigated 1 - 4 ML ultrathin films of NaCl epitaxially grown on different copper substrates with low temperature scanning tunneling microscopy. The ultrathin NaCl films have been used as substrate to investigate the adsorption of single metal atoms and metal dimers. The binding sites of Cu, Ag, Au and Pd were determined, local spectroscopy performed and atomic manipulation processes were studied.

Most importantly on a polar insulating surface, the charge state of individual adatoms can be controlled. An exemplary system are single Au adatoms adsorbed on a 2ML thick NaCl film on Cu(111). By positioning the STM tip above an Au adatom and applying a voltage pulse, the adatom can be reversibly switched between its neutral and negatively charged state. Most importantly, both states are stable, that is, an additional charge remains on the adsorbate until it is removed by a voltage pulse of reversed sign. Two different experimental observations suggest that an Au adatom in its original state is neutral, whereas after manipulation it is negatively charged. In the first experiment, the scattering of electrons in the interface state (IS) band of NaCl/Cu(111) was used as a probe to characterize the two different states of the Au adatoms. While no scattering was observed in the original (neutral) state, the manipulated (charged) state shows a strong scattering due to the long range potential of a charged ion. In the second experiment, the Au adatom charge state was probed by the direct interaction of the adatom with a negatively biased tip. When applying a bias voltage of about 1 V and laterally approaching the tip to the position of a manipulated(charged) Au adatom, the adatom moved away from the negatively biased tip. The experimental results are supported by density functional calculations by F. Olsson and M. Persson, which have shown that the Au anion state is stabilized by a large outward relaxation of the Na due to the high ionic polarizability of the insulating film. This simple physical mechanism behind the charge bistability suggests that this is a common phenomenon for adsorbates on polar insulating films

