Single-molecule charging investigated by nc-AFM

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The striving towards miniaturization in the electronics industry seeks the ultimate goal to build devices comprising single atoms or molecules as the only active components. Design and application of such devices will require a high degree of charge localization within the involved atoms or molecules while maintaining precise control over the occupation of their electronic levels. These needs have motivated numerous studies in experimental physics on fundamental aspect of electronic decoupling of nanostructures as well as determination and - more importantly - manipulation of their charge state. When investigating single atoms or molecules, scanning probe microscopy (SPM) is the tool of choice, as its ability to image and manipulate at the atomic scale is unmatched. Indeed, scanning tunneling microscopy (STM) has proven its potential to determine and change charge states of atoms and molecules [1, 2]. Non-contact atomic force microscopy (nc-AFM) has achieved the determination of charge states as well [3]. However, the dynamic response of the AFM due to charging events has been measured so far mainly for large, many-electron systems such as carbon nanotubes [4] or semiconducting quantum dots [5].

Using low-temperature tuning-fork-based nc-AFM/STM, we show that charging and discharging events of single molecules yield clear fingerprints in the dynamic response of the measured frequency shift (*df*). The charge-transfer complex TCNQ-TMTTF deposited onto Au(111) forms a double-barrier-tunneling-junction in proximity of an SPM tip, allowing for gating the charge state of the TCNQ molecules by sweeping the applied bias voltage [6]. The simultaneously measured *df* signal shows a pronounced dip in its parabolic response at the charging/discharging bias. Detailed analysis reveals that the dip marks the transition from one force parabola to another as adding or removing a single electron to or from the molecule causes a sudden shift in the electrostatic forces. This result and further experiments demonstrate the capability of nc-AFM to detect single-electron processes in molecules.

References

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