Probing charge-state, spin and vibrational excitations of radical molecules by low-temperature scanning probe microscopy

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Achieving the precise control over the charge-state of molecules occupied by a single electron (spin) is crucial for their future implementations in quantum information platforms. In this contribution, I will show the self-assembly on Ag(111) of tetrabromotetraazapyrene (TBTAP) molecules using scanning tunneling microscopy (STM) and atomic force microscopy (AFM) [1]. Due to their strong electron-accepting character, TBTAP molecules are able to trap a single electron transferred from the substrate in an unoccupied molecular orbital. This electron induces a 1/2 molecular spin state confirmed by the detection of a Kondo resonance in tunneling spectroscopy (dI/dV). Using the local electric field of the tip, we are able to control the transition from the anionic ($TBTAP^-$) to its neutral ($TBTAP^0$) counterpart. This transition leads to vibrational excitation of the molecule (Figure 1), which is experimentally reflected by a series of peaks in the dI/dV spectra. By transferring this precursor to a superconducting lead surface [2], I will also show that anionic TBTAP molecules can form an extended two-dimensional electron spin lattice which carries topological edge states.

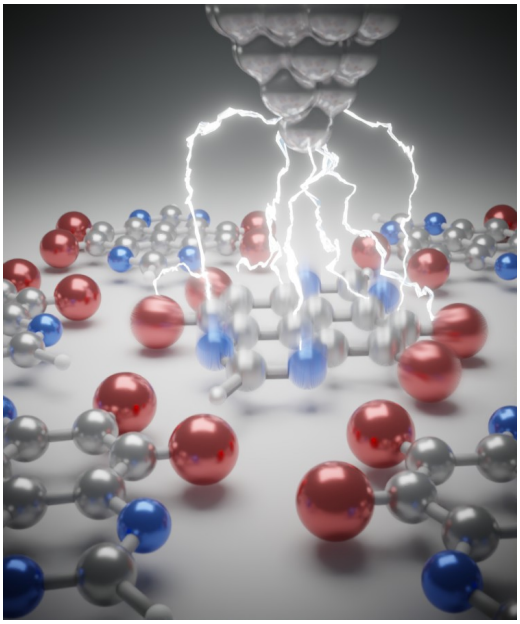

Fig. 1. Illustration of the vibrational excitation of a single molecule following its discharge triggered by the electric field of a STM tip.

[1] Li, C.; Kaspar, C., Zhou, P.; Liu, J.-C; .Chahib, O.; Glatzel,T.; Häner, R.; Aschauer, U.; Decurtins, S.; Liu, S.-X.; Thoss, M.; Meyer, E.; Pawlak, R. *Nat. Comm.*, in review.

[2] J.-C. Liu, C. Li, H. Chen, C. Drechsel, P. Zhou, R. Häner, U. Aschauer, T. Glatzel, S. Decurtins, S.-X. Liu, D. Loss, J. Klinovaja, W. Wulfhekel, E. Meyer, R. Pawlak. Submitted.