Accessing non-equilibrium states at atomic scales

J. Repp [1]

[1] *Department of Physics, University of Regensburg, 93040 Regensburg, Germany*

Scanning probe microscopy (SPM) has revolutionized our understanding of the atomistic world. Conventional SPM, however, is an inherently slow technique – too slow to capture transition states in excitation processes in most cases. While ultra-fast non-equilibrium phenomena is enabled by terahertz (THz) scanning tunneling microscopy (STM) [1], another approach gives us access to intermediate timescales that are relevant for spin precession and relaxations. We introduce a novel variant of SPM by combining principles of STM and atomic force microscopy (AFM). Instead of the usual direct current in conventional STM, we drive a tiny alternating current between the microscope's tip and a single molecule under study. We exploit the single-electron sensitivity of AFM [2] in detecting the current which consists of only a single electron per AFM-cantilever oscillation cycle, tunneling back and forth between tip and molecule. This enables operation in absence of any conductance of the underlying substrate, while retaining the capability of imaging electronic states with sub-angstrom resolution. Thereby, we can access out-of-equilibrium charge states that are out of reach for conventional STM [3]. Extending this technique by electronic pump-probe spectroscopy [4], see Fig. 1, we measured the triplet lifetime of an individual pentacene molecule on an insulating surface [5] and lifetime quenching by nearby oxygen molecules. Combined with radio-frequency magnetic-field driving we introduce AFM-based electron spin resonance and spin manipulation showing long spin coherence in single molecules [6].

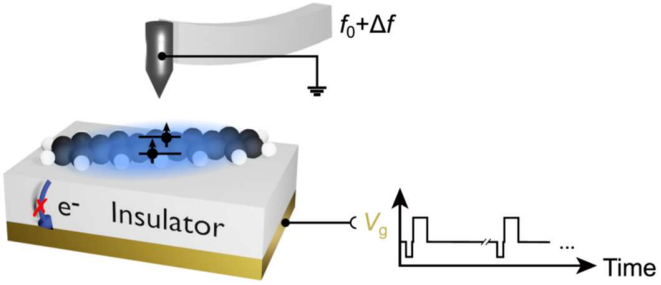

Fig. 1: *Schematic of the setup, by which we probe the triplet lifetime of an individual pentacene molecule. The molecule is placed on an insulator preventing any electron exchange with the substrate. By means of voltage pulses we bring the molecule into its triplet state and probe how fast the triplet decays.*

References:

[1] T. L. Cocker et al., Nature **539**, 263 (2016).
[2] J. Klein, C. C. Williams, Appl. Phys. Lett. **79**, 1828 (2001).
[3] L. L. Patera et al., Nature **566**, 245 (2019).
[4] S. Loth et al., Science **329**, 1628 (2010).
[5] J. Peng et al., Science **373**, 452 (2021).
[6] L. Sellies et al., arXiv:2212.12244 (2022).