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SEMINAIRE IPCMS

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à 15h à l'auditorium

« Accessing non-equilibrium states at the atomic scale »

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Accessing non-equilibrium states at the atomic scale

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Scanning probe microscopy has revolutionized our understanding of the atomistic world. It is, however, is an inherently slow technique – too slow to capture transition states in excitation processes in most cases. Two complementary approaches that allow accessing non-equilibrium phenomena with scanning probe microscopy will be presented, opening a new arena for atomistic studies.

We introduce a novel variant of scanning probe microscopy by combining principles of scanning tunneling (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 [1] 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 Angstrom resolution. Thereby, we can access out-of-equilibrium charge states that are out of reach for conventional STM. Our results unveil the effects of electron-transfer and polaron formation on the single-orbital scale [2,3].

Accessing ultra-fast non-equilibrium phenomena is enabled by terahertz (THz) scanning tunneling microscopy [4] (THz-STM) through combining STM with lightwave electronics. In THz-STM, the electric field of a phase-stable single-cycle THz waveform acts as a transient bias voltage across an STM junction. These voltage transients [5] may result in a net current that can be detected by time-integrating electronics. The recent development of this lightwave STM has enabled the combined femtosecond and sub-angstrom resolution in observing matter [6] (see Fig. 1).

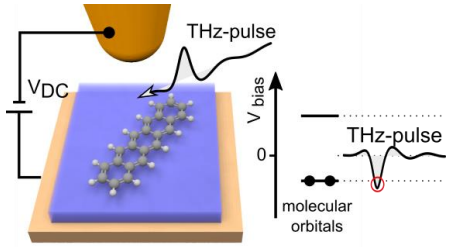

Fig. 1 | In THz-STM, the electric field of a phase-stable single-cycle THz waveform acts as a transient bias voltage. Such transients allow observing and even controlling individual molecules directly on the relevant ultrafast and ultrasmall scales.

We demonstrate the first combined femtosecond and sub-angstrom access in the control of matter. Ultrafast localized electric fields in lightwave STM enable exerting atom-scale femtosecond forces to selected atoms. By means of these atomic forces on the intrinsic timescale of molecules, coherent atomic motion can now be excited. Utilizing this coherent structural dynamics, we can modulate the quantum transitions of a single-molecule switch by up to 39%. We directly visualize the coherent excitation of the switch in a real-space femtosecond single-molecule movie [7].

References:

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