Physikalisches

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Snacks und Getränke im Anschluss

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Actuating and probing a single-molecule switch at femtosecond timescales

Accessing ultra-fast non-equilibrium phenomena is enabled by terahertz (THz) scanning tunneling microscopy [1] (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 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 [2]. We now demonstrate the first combined femtosecond and subangstrom access in the control of matter. Ultrafast localized electric fields in lightwave STM enable exerting atom-scale femtosecond forces to selected atoms. By shaping atomic forces on the intrinsic timescale of molecules, coherent atomic motion can now be excited. Utilizing this coherent structural dynamics, we can modulate the guantum transitions of a single-molecule switch by up to 39%. We directly visualize the coherent excitation of the switch in the first femtosecond single-molecule movie [3]. To resolve the impact of coherent control of the single-molecule switch, alongside, we introduce single-shot action spectroscopy in lightwave STM as the first concept resolving individual path-selective reaction events of a single molecule in space and time. With this novel concept, we detect the outcome of every single laser shot and further separate the statistics of the two inverse reaction paths. Our results open a new chapter in the control and observation of reactions of individual molecules directly on the relevant ultrafast and ultrasmall scales.

[1] T. L. Cocker et al., Nature Photon. 7, 620 (2013).

[2] T. L. Cocker et al., Nature 539, 263 (2016).

[3] D. Peller et al., in preparation.



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