Séminaire AXE 1 - Sciences et Matériaux Quantiques

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Ultrafast dynamics and spectroscopy of solid surfaces using light-coupled STM

The macroscopic physical properties of solids are governed by the complex interplay between their internal – charge, lattice, spin and orbital - degrees of freedom. Conventional optical spectroscopy allows the study of the optical properties of solids in the steady-state, while ultrafast pump-probe approaches provide unique insights into the microscopic interactions and couplings that determine their non-equilibrium response and govern their functionality. Despite the unprecedented simultaneous energy, momentum and time resolution offered by established optical and ultrafast surface science techniques, their spatial resolution is limited to tens to hundreds of nanometers. In the presence of (sub-) nanoscale spatial inhomogeneities, important local information is thus lost, although it can be crucial to fully understand and tailor the properties of solid matter.

Recent advances in coupling broadband optical and terahertz (THz) radiation with low temperature scanning tunneling microscopes (STM) have greatly expanded the possibilities to study the ultrafast dynamics and optical properties of surfaces at the atomic scale [1]. In this talk, I will discuss our recent efforts to apply ultrafast all-optical and THz-lightwave-driven STM (THz-STM) [2] as well as local optical spectroscopy to study the ultrafast dynamics and light emission and scattering properties of complex solid surfaces. First, I will discuss the role of the local electronic structure of ultrathin ZnO films on Ag(111) for tip-enhanced Raman spectroscopy (TERS) [3], for tunneling-induced plasmonic light emission [4], and for femtosecond photocurrent generation [5] from these films, and will demonstrate that the latter allows the local detection of coherent phonons by all-optical ultrafast STM [5]. Second, I will show that THz-STM allows to probe the coherent dynamics of the charge density wave (CDW) amplitude mode in the layered quantum material 1T-TaS2, and discuss how the coherent lattice motion appears as a transient modulation of the local density of states (LDOS) probed by STM [6]. Finally, I will show and discuss the ability of THz-STM to probe and image photoexcited dynamics on time scales much faster than a single THz cycle [6].

References:
[1] M. Müller, Prog. Surf. Sci. 100727, in press (2023)
[2] T. Cocker et al., Nat. Phot. 15, 558-569 (2021)
[3] S. Liu et al., Nano Lett. 19, 8 (2019)
[4] H. Wiedenhaupt et al., in preparation
[5] S. Liu et al., Sci. Adv. 8, 42 (2022)
[6] L. Parra Lopéz et al., in preparation

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