Reconstruction of effective THz fields for probing structural dynamics on metal catalyst surfaces

The design of effective catalysts requires a detailed microscopic understanding of the chemical reaction dynamics [1] which is on timescales ranging from a few femto- to several picosenconds. Low-frequency vibrational modes in the THz-range are mostly responsible for the molecule-substrate interaction [2]. Hence, intense THz radiation can be used to excite an ensemble of adsorbate molecules resonantly into a coherent motion [3] and can in turn be studied by suitable probing techniques, e.g. X-ray photoelectron diffraction (XPD) owing to the large cross-section of the electrons and high temporal resolution [4,5]. The orientation and amplitude of the vibrational modes is controlled by the character of the exciting electric field in close proximity to the surface. An experimental access to the effective field, a superposition of incoming and reflected laser pulse, is thus highly desirable. In this study we present a way to reconstruct the effective electric fields generated by THz-light pulses incident on metallic and dielectric surfaces by means of photoelectron streaking, i.e. the momentum gain of photoexcited electrons in a dressing laser field. Furthermore, we present data from a THz-pump/ extreme ultraviolet (XUV)-probe experiments on nanostructured metallic thin-films which were performed at the free electron laser facility FLASH at DESY (Hamburg). Here, XUV-emitted photoelectrons were subject to streaking by a strong, few cycle THz pulse. Our 2D electron analyzer, capable of probing both the kinetic energy and emission angle of the photoelectrons, allowed us to follow the change in the electron momentum distribution at different delays between THz and XUV pulse (Fig 1. (a)). A representative complete delay trace obtained on a Pt(111) thin film sample is depicted in Fig 1. (b). The components parallel and perpendicular to the surface lead to distinct changes in the momentum distribution and enable us to reconstruct the amplitude and phase of the effective THz field (Fig 1. (c)). We compare the results on the nanostructured thin-films with data obtained on a Pt(111) bulk crystal.

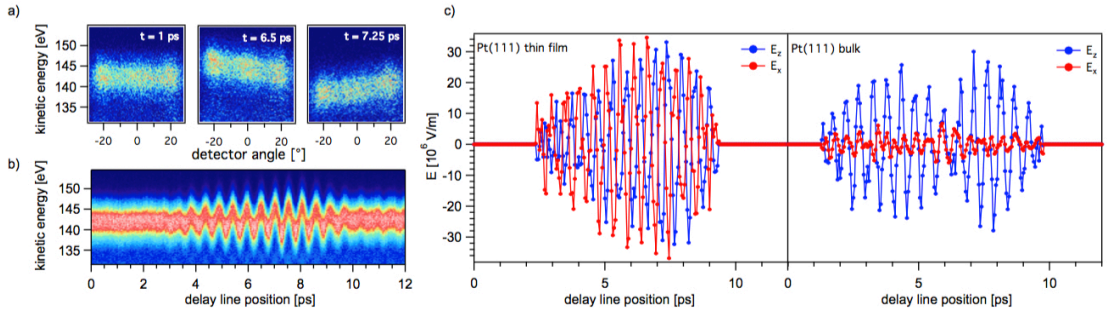

Fig. 1: (a) Photoelectron momentum distribution as detected in our 2D electron analyzer at different delay positions between THz and XUV pulse. (b) Angle- integrated THz streaking trace of the Pt(111) thin film valence band. (c) Reconstructed parallel (E_X) and perpendicular (E_Z) components of the THz pulse at a Pt(111) thin-film (left) and bulk (right) surface.

References

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