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Strong-field-induced attosecond dynamics in SiO_2

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Abstract. Striking field-induced changes in the absorption near the Si L-edge of SiO_2 exposed to a near-infrared laser field of several V/Å delivered by a few-cycle pulse are observed with sub-100 attosecond extreme ultraviolet pulses by means of attosecond transient absorption.

1 Introduction

Understanding the behavior of dielectric materials under the influence of strong electric fields is of fundamental interest, but, unlike in gaseous media, direct access to the ionization dynamics via electron and ion detection is impeded inside a solid. As the damage threshold of dielectrics increases with decreasing exposure time, ultrashort laser pulses of only a few-cycle duration allow for exposure of dielectrics to electric fields of several V/Å without damage and provide insight into dynamics on ultrashort time scales [1,2]. For electric field strengths close to the critical field strength defined by a change in electron potential energy on the order of the size of the bandgap E_g over the lattice period *a*, Zener-type transitions from the valence band into the entire conduction band become possible. Due to the large spectral width of the conduction band of SiO_2 of about 10 eV, the induced polarization dynamics are on the 100-attosecond (as, 1 as = 10^{-18} s) time scale. Using extreme ultraviolet (XUV) attosecond pulses to excite 2*p* electrons of Si to conduction-band states allows probing the sub-cycle response of the conduction band states to a strong near-infrared laser field.

2 Experimental methods

In our experiment, we use ultra-broadband, isolated attosecond pulses in the extreme ultraviolet spectral domain with energies ranging from ~90 eV to ~135 eV produced using high harmonic generation (HHG) in a neon gas target from few-cycle visible-near-infrared (NIR) laser pulses with stabilized carrier-envelope-phase and 400-μJ pulse energy (FemtoPower Compact Pro, Femtolasers). Taking advantage of the much smaller divergence of the XUV radiation, the collinearly propagating laser and XUV beams are separated into two arms of a Mach-Zehnder-type interferometer by means of a perforated mirror, which reflects the optical and transmits the XUV pulses [3]. In the XUV arm of the interferometer three subsequent Rhodium coated mirrors under 75 degrees provide an overall

reflectance >45 percent between 90 eV and 120 eV, allowing for both high flux and large bandwidth beyond the limits of conventional XUV multilayer optics. The laser beam is re-collimated in the optical arm, and its intensity can be controlled via a variable aperture. The length of the optical arm of the interferometer and therefore the timing between the two arms can be varied in sub-micrometer steps via a mirror mounted on a high resolution piezo linear stage. The XUV and the laser beam are recombined at a second perforated mirror and focused with a toroidal mirror (Zeiss) on the free-standing chemical-vapor-deposited 125 nm thick SiO_2 sample. The focal spot size on target is 125 μm for the laser beam and 40 μm for the attosecond XUV beam. In order to prevent lateral intensity averaging, we observe the XUV transmission on the optical axis of the laser beam. The XUV beam transmitted through the sample is spectrally dispersed by a flat-field grating (Hitachi) and projected on a XUV-sensitized camera. Introducing a 0.2 mm-wide slit between the target and the grating provides a spectral resolution of ~350 meV.

XUV spectra after transmission through the SiO_2 sample are recorded with 100-as step size in delay between the XUV pulse and the laser pulse and divided by a spectrum recorded without transmission through the sample. The laser field strength impinging on the thin film is characterized by introducing a dilute neon gas target in front of the SiO_2 sample and recording an attosecond streaking spectrogram, shown in Fig. 1, at each delay step in addition to the transmission measurement. Processing spectrograms with a frequency-resolved optical gating algorithm (ATTOgram [4]) allows us not only to reconstruct the waveform of the laser field, but also to determine the XUV pulse duration being as little as 72 as.

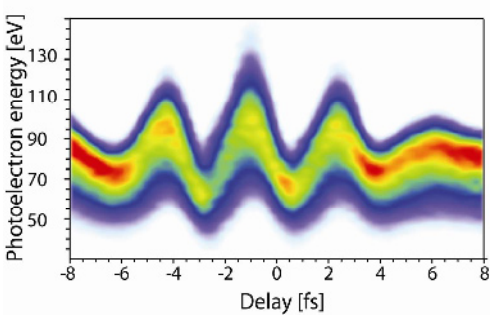

Fig. 1. Attosecond streaking spectrogram of Ne*2p* recorded simultaneously with the transient absorption spectrogram.

3 Results and discussion

The XUV attosecond pulse absorption of a thin dielectric film (fused silica) irradiated with a waveform-controlled few-cycle NIR laser pulse measured as a function of the delay between the two pulses reveals a sub-cycle build-up of transient absorption bleaching at the SiO_2 L-edge absorption and an attosecond response to the oscillating field. We evaluate the optical density $OD(\hbar\omega_{XUV})=\alpha(\hbar\omega_{XUV})d$, with α being the absorption coefficient of fused silica at the XUV photon energy of 109 eV integrated over a 1-eV bandwidth at the peak of L-edge-conduction band absorption, shown in the upper panel of Fig. 2 (a) (solid line). The strong modification of up to 10% in the sample's transmittivity ends abruptly after several femtoseconds, coinciding with the end of overlap with the driving field. Simultaneous attosecond streaking in neon allows us to directly link the absorption dynamics to the experimental field, shown in the lower panel of Fig. 2 (a) and (b), with a field strength of up to ~2 V/Å. Also the energetic position of the absorption line, evaluated with peak fitting, shows pronounced oscillations with twice the optical field frequency (Fig. 2 (b), solid line in upper panel), demonstrating for the first time the observation of an instantaneous Stark-shift in condensed matter at optical frequencies. Error bars in Fig. 2 (b) correspond to the 95% confidence intervals of the least-squares-fit.

To describe the polarization dynamics leading to a modification of the XUV absorption, a 1D-quantum mechanical model similar to [5] is proposed based on the time-dependent Schrödinger equation. Its results are depicted as dashed lines in Fig. 2(b) and show excellent agreement with the experimental data. Using the experimental electric field as input for the calculation, the phase of the modulation present in the experimental response relative to the driving field can be traced back to a strong induced polarizability *inside* the solid.

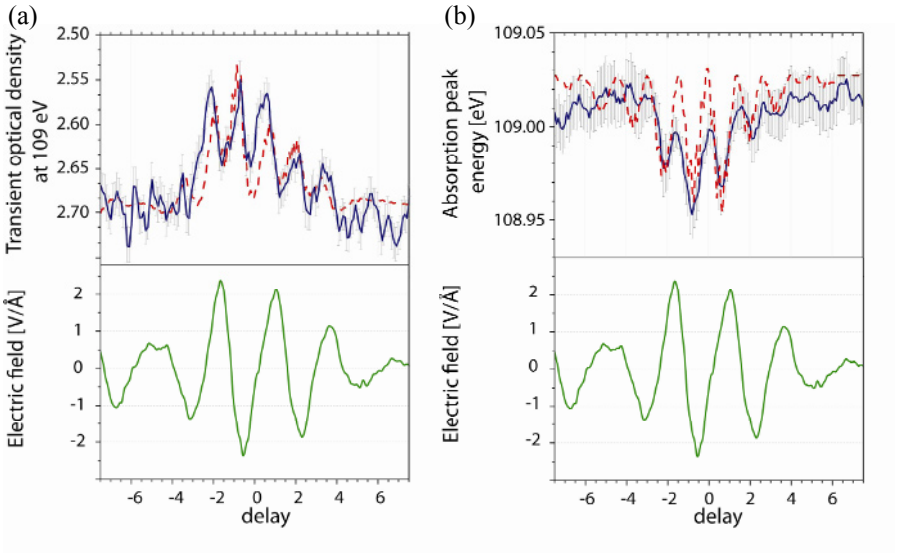

Fig. 2. The change in strength and energetic position of the main absorption line at 109 eV is evaluated in the upper panels of (a) and (b), respectively (solid lines), and compared with theoretical results (dashed lines). The timing relative to the NIR driving laser field (lower panel in (a) and (b)) is provided by simultaneous attosecond streaking spectroscopy in neon.

4 Conclusions

The powerful combination of attosecond transient absorption spectroscopy with attosecond streaking characterization of the instantaneous field applied to a thin dielectric sample reveals transient absorption bleaching of attosecond XUV pulses during the overlap with a several-V/Å waveform-controlled NIR driving laser field exhibiting sub-cycle modulation following the instantaneous field. While optical-field-driven effects at surfaces have been observed earlier [6], our work marks the first attosecond observation of processes *inside* a solid and demonstrates the feasibility of controlling the electronic properties of dielectrics reversibly on a sub-femtosecond timescale with the electric field of light.

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