Tailoring of High-Field Multi-THz Waveforms with Sub-Cycle Precision

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Abstract: Shaping of extremely intense mid-infrared transients by means of time-domain slicing and frequency-domain synthesis is demonstrated. We achieve phase-stable transients with multiple MV/cm peak fields and having single-cycle duration and strong polar asymmetry.
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We present two alternative approaches to control duration and optical cycles of multi-THz waveforms. The first approach relies on waveform slicing directly in the time domain by means of an ultrafast semiconductor switch [1]. In contrast to manipulation in the frequency domain, this method inherently increases the spectral bandwidth by generation of new spectral components. We demonstrate for the first time sub-cycle slicing for the generation of high-field and phase-locked multi-THz transients [2] containing only few cycles of light. The second approach is based on coherent ω-2ω synthesis resulting in high-field multi-THz waveforms with strong polar asymmetry. To this end, we generate broadband second harmonic of the few-cycle fundamental waveform leading to an octave-spanning phase-locked THz spectrum. By controlling the phase difference between the first and second harmonics, we demonstrate fully tunable polar asymmetry of the synthesized THz transients.

The ultrafast time-domain slicing is achieved by a controlled reflection of intense multi-THz pulses from an intrinsic germanium (i-Ge) wafer placed at Brewster's angle. The reflected THz field is detected as a function of electro-optic sampling (EOS) delay time t_1. The ultrafast switching is triggered by 18-fs-short pulses with a center wavelength of 1080 nm and energies up to 15 μJ. We employ a two-stage non-collinear optical parametric amplifier [3] in order to achieve intense broadband control pulses necessary for an efficient switching process. The temporal offset t_2 of the control pulse with respect to the THz transient is adjusted with a delay stage. By adjusting the delay time t_2, we are able to precisely control the temporal position of the leading edge and hence the pulse duration of the THz transient. This is illustrated in Fig. 1(a) which shows the reference high-field multi-THz waveform containing about 2.5 oscillation cycles and the waveform as sliced by the control pulse with the fluence of 13.7 mJ/cm^2. The resulting THz transient contains only a single optical cycle and has a remarkably high peak electric field of about 10 MV/cm. The generation of new spectral components results in nearly doubling the bandwidth of the sliced transient as can be clearly seen in Fig. 1(b). Fig. 1(c) shows the reflected THz field at the maximum and the minimum of the reference waveform as a function of the delay time t_2. The obtained reflectivity onset time of 45 fs is smaller than the period of the multi-THz oscillation indicating slicing operations in the sub-cycle range.

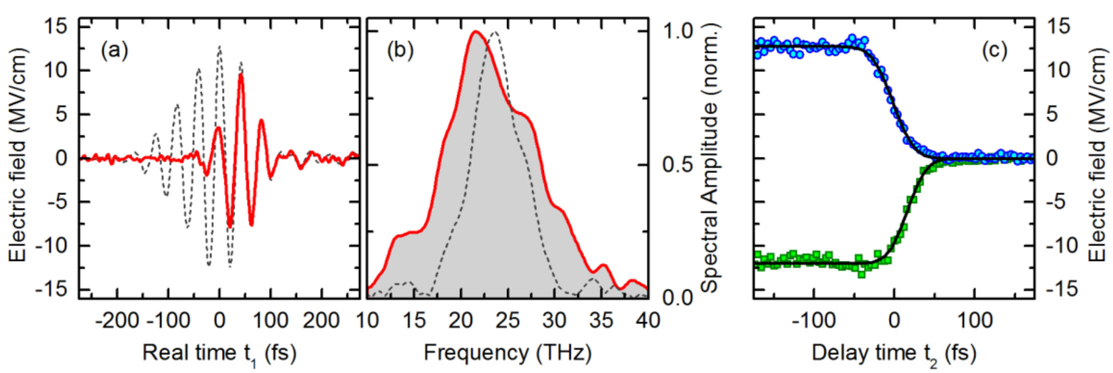

Fig. 1. (a) Time-domain profiles of multi-THz transients. The dashed line corresponds to the reference transient fully reflected by the photoexcited Ge wafer. The solid red line shows the sliced single-cycle THz waveform. (b) Corresponding normalized amplitude spectra of the reference (dashed line) and the sliced (solid line) waveforms indicate a notable spectral broadening produced by the sub-cycle switching. (c) Dynamics of the slicing process illustrated by electric fields measured at a fixed EOS time t_1 as a function of the delay time t_2. The evolution of the reflected field maximum (circles) and the adjacent minimum (squares) is shown together with fitting functions (solid lines).

In the second frequency-domain approach, we combine phase-locked fundamental and collinearly-generated second harmonic of the THz transient to synthesize a novel functional waveform with controlled polar asymmetry of the temporal envelope. Such asymmetry is directly achieved by the change of the relative phase of the injected second harmonic with respect to the fundamental wave. The asymmetry allows for coherent control of ultrafast transport in condensed matter [4], as well as sub-cycle control of XUV and THz generation mechanisms in plasmas [5,6]. For generation of the synthetic fields we use intermediate focusing plane between the generation and detection. The second harmonic is obtained by placing $AgGaSe_2$ nonlinear crystal ($\theta = 56°$, $\varphi = 45°$, thickness of 350 µm) in the intermediate focus (60 µm FWHM) of the fundamental phase-locked transient. The emerging electric fields of the fundamental and second harmonic are recollimated and transmitted through a dispersive element (500-µm-thick InAs wafer), which serves as phase plate for control of the relative phase of the fundamental and second harmonic, and hence the asymmetry of the synthesized waveform. The resulting THz field is resolved by means of the EOS. Fig. 2 shows the waveform for the condition of π relative phase shift, resulting in largest (positive) asymmetry in the envelope. Strong envelope asymmetry is clearly visible in the time domain, due to the efficient second harmonic generation process (49% field yield).

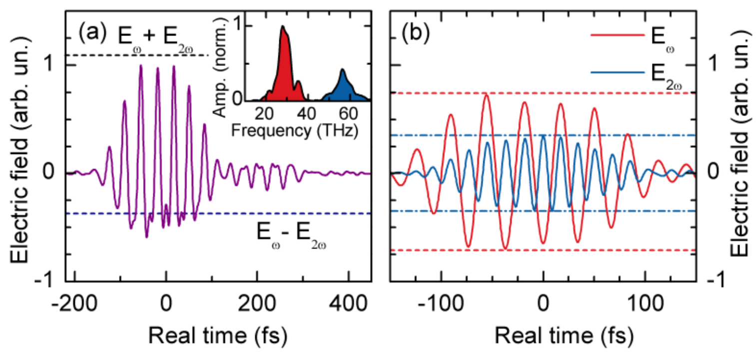

Fig. 2. (a) Time-domain profile of synthetic two-color THz waveform. Strong envelope asymmetry is clearly visible by comparison of the shaded areas; Fourier components of the transient in (a) are depicted in the inset; (b) Decomposition of the spectrum into the fundamental (red) and second harmonic (blue) transients allows determination of the maximal field amplitude of each component (E_ω, $E_{2\omega}$, respectively). Comparison of the maximum (E_ω+$E_{2\omega}$) and minimum (E_ω-$E_{2\omega}$) possible field values (dotted lines in panel a) are in very good agreement with the observed asymmetry in the synthesized waveform.

In conclusion, we have demonstrated time-domain slicing based on an ultrafast semiconductor mirror which enables the control ultrashort optical waveforms on a sub-cycle level of precision. Using this technique, we succeeded in generation of single-cycle multi-THz transients with a peak electric field above 10 MV/cm. This approach can be scaled up for even higher THz fields and extended by implementing an additional "switch-off" device through gated transmission in Ge for arbitrary tailoring of multi-THz waveforms. In the frequency domain approach, we generated and detected synthetic THz transients with strong and controlled polar asymmetry. Such transients can be used for novel field-controlled transport and light-matter interaction experiments, particularly accessing the non-perturbative regimes.

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