Phase-locked Multi-THz High-Harmonic Generation by Dynamical Bloch Oscillations in Bulk Semiconductors

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Abstract: Ultra-intense and CEP-stable waveforms in the multi-THz range control dynamical Bloch oscillations and interband polarization in bulk GaSe, leading to the emission of all-coherent high-order harmonics covering 12.7 optical octaves from THz to VIS regimes.
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Bloch oscillations are among the most spectacular quantum manifestations of electrons in crystalline solids. When an electric field accelerates an electron, its wavelength shortens. Once the latter equals twice the lattice constant, the wave should undergo Bragg reflection, causing electrons to oscillate in reciprocal and real space [1,2]. Ultrafast carrier scattering and dielectric breakdown under constant-field biasing have hampered the experimental observation of this long-standing prediction in bulk crystals [2]. Recently, high harmonic generation has been attributed to a dynamical version of Bloch oscillations [3]. Controlling the precise shape of the optical fields, however, has been out of reach due to the fluctuating carrier-envelope phase (CEP) of the laser pulses. Novel developments in field-resolved multi-terahertz optics provide low-frequency CEP-stable electromagnetic waveforms, which serve as a sub-cycle bias for high-field experiments [4,5]. Here, we employ phase-locked multi-THz fields as high as 72 MV/cm to control all-coherent charge transport in gallium selenide (GaSe) on femtosecond timescales. Electric fields comparable to atomic potential gradients off-resonantly drive coherent interband polarization and dynamical Bloch oscillations, resulting in the emission of phase-stable high-order harmonics (HH) covering the frequency range from 0.1 to 675 THz. The observed spectral width corresponds to 12.7 optical octaves and sets a new record for phase-stable femtosecond pulses [6]. Quantum interference of different excitation pathways allows for sensitive control of the HH generation process via the CEP of the driving waveform.

We generate few-cycle multi-THz transients with adjustable CEP and frequency via difference frequency mixing [4]. Waveforms featuring a center frequency of 30 THz and peak electric fields of 72 MV/cm (Fig. 1a) are incident on a 220 µm thick sample of undoped bulk GaSe. Electro-optic detection of the transmitted waveform with 8-fs near-infrared gate pulses reveals low frequency components between 0.1 and 10 THz, stemming from optical rectification, as well as fast oscillations at a frequency of 60 THz, corresponding to the second harmonic of the fundamental wave (Fig. 1b). Intriguingly, ultrabroadband traces also exhibit spectral signatures at the third and fourth harmonics (Fig. 1c, blue curve). Switching to indium gallium arsenide and silicon detectors, we observe a plateau-like region of high-order harmonics. Even and odd harmonic orders feature similar intensities up to the bandgap of GaSe at a frequency of 476 THz where the photoluminescence peak nearly coincides with the 16th harmonic. Beyond the fundamental energy gap, the harmonic intensity decreases due to interband absorption and is detected up to the 22nd order. Remarkably, all spectral components are absolutely phase-stable, as verified by electro-optic sampling of the lowest orders (Fig. 1b) and f-2f-interferometry for higher orders (not shown). These findings indicate an all-coherent generation mechanism for the emission of high-harmonics. The scaling of HH intensities with the peak driving field E reveals the non-perturbative character of the dynamics: While the nth order intensity I_n follows $I_n \sim E^{2n}$ for low fields, it approaches $I_n \sim E$, asymptotically, for large fields (not shown) [6].

High-harmonic generation in atoms has been explained by a semiclassical three step model including ionization, acceleration and recollision [7]. In solids, however, the quantum mechanical wave nature of electrons dominates the physics of high-field transport. We extend the many-body theory of Ref. 8 to a five band model, including inter- and intraband excitation as well as THz-induced band mixing. Figure 1d shows the calculated dynamics of the electron density n_e in the first conduction band for the experimental driving waveform and internal peak fields of 14 MV/cm. Carriers are dominantly injected at every second field maximum and simultaneously accelerated along the direction of the electric field. The electron wave packet starts to oscillate following the external field and, once the Brillouin zone boundary at the K-point is reached, enters the Brillouin zone from the opposite side again. For highest fields, multiple such Bragg reflections occur during one half cycle of the driving field, giving rise to HH emission.

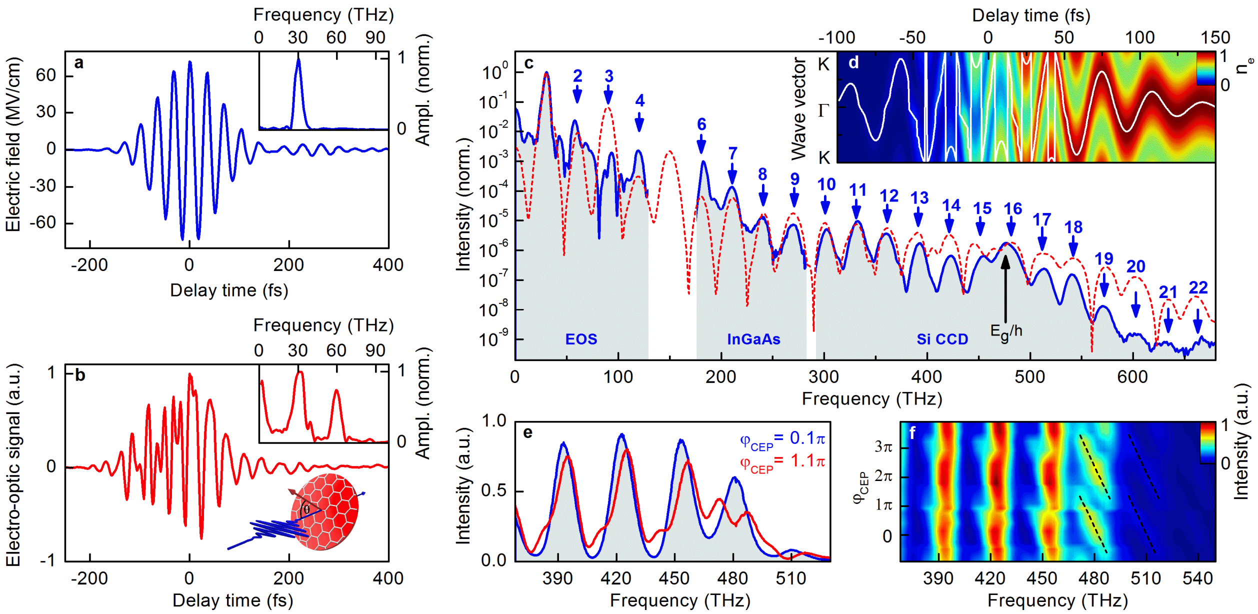

Fig. 1: a Electro-optically detected waveform of the multi-THz driving field featuring an amplitude of 72 MV/cm in air and a central frequency of 30 THz (inset: corresponding spectrum). **b** Electromagnetic transient generated in a 220 µm thick GaSe sample (θ = 70°) by the pulse shown in **a**, revealing a superposition of fundamental, optical rectification and second harmonic components (insets: corresponding spectrum and experimental geometry defining the angle of incidence θ). **c** Ultrabroadband phase-stable HH intensity spectrum generated in GaSe (220 µm, θ = 50°) by the waveform shown in **a** as detected by electro-optic sampling (EOS), an InGaAs and a Si detector array (blue curve), respectively. The red dashed line shows the calculated HH intensity obtained by the quantum-mechanical five-band model. E_g marks the band gap energy of GaSe. **d** Computed dynamics of the electron distribution n_e in the first conduction band for the experimental waveform and internal peak fields of 14 MV/cm (colormap). The white line highlights the center of the electron wave packet. **e** Measured HH intensity spectra generated by driving waveforms with carrier-envelope phases of $\varphi_{CEP} = 0.1\pi$ (blue) and 1.1π (red). **f** Systematic dependence of HH spectra on the CEP of the terahertz transient. HH peaks of order $n \geq 15$ shift in frequency with a slope of -2.5 THz rad^{-1} (indicated by the black dashed line).

Our calculations reproduce the measured HH intensities very well (red dashed curve in Fig. 1c) and identify dynamical Bloch oscillations along with coherent interband excitation as the origin of HH emission. Due to quantum interference of multiple excitation pathways involving different electronic bands, HH generation is expected to depend sensitively on the amplitude and phase of the THz waveform. In our experiment, we vary the CEP of the driving field and record HH spectra. A pure sign flip of the transient already radically alters the shape and intensity of the emitted spectra (Fig. 1e). For $\varphi_{CEP} \approx 0$, a sinusoidal modulation of the spectrum is observed, while additional peaks spaced by 15 THz appear for $\varphi_{CEP} \approx \pi$. For a comprehensive picture, we record intensity spectra while varying φ_{CEP} continuously from $-\pi$ to 4π (Fig. 1f). High-harmonic generation below 400 THz is only slightly affected, whereas the intensity maximum of the 16th harmonic shifts about a frequency of 480 THz, with a slope of -2.5 THz rad^{-1} (black line in Fig. 1f). These features are well reproduced by our full quantum mechanical theory [6].

In conclusion, coherent interband excitation and intraband acceleration in a bulk semiconductor are off-resonantly driven by intense, phase-stable multi-THz waveforms. Precise tuning of the CEP of the driving fields allows for sensitive control of the carrier dynamics, which lead to the emission of an all-coherent HH spectrum covering 12.7 optical octaves from THz to visible spectral ranges. The results highlight quantum phenomena relevant for future semiconductor devices at terahertz clock rates and open the door to a novel regime of high-field transport on timescales shorter than a single oscillation cycle of light.

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