Sub-cycle control of terahertz high-harmonic generation by dynamical Bloch oscillations

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Ultrafast charge transport in strongly biased semiconductors is at the heart of high-speed electronics, electro-optics and fundamental solid-state physics¹⁻¹³. Intense light pulses in the terahertz spectral range have opened fascinating vistas¹⁴⁻²¹. Because terahertz photon energies are far below typical electronic interband resonances, a stable electromagnetic waveform may serve as a precisely adjustable bias⁵,¹¹,¹⁷,¹⁹. Novel quantum phenomena have been anticipated for terahertz amplitudes, reaching atomic field strengths⁸⁻¹⁰. We exploit controlled (multi-)terahertz waveforms with peak fields of 72 MV cm⁻¹ to drive coherent interband polarization combined with dynamical Bloch oscillations in semiconducting gallium selenide. These dynamics entail the emission of phase-stable high-harmonic transients, covering the entire terahertz-to-visible spectral domain between 0.1 and 675 THz. Quantum interference of different ionization paths of accelerated charge carriers is controlled via the waveform of the driving field and explained by a quantum theory of inter- and intraband dynamics. Our results pave the way towards all-coherent terahertz-rate electronics.

Once called the ‘terahertz gap’, the spectral boundary between electronics and optics has recently been made accessible with unique photonics tools¹⁹. Broadband terahertz pulses with inherently stable carrier-envelope phase (CEP) and peak electric fields of 1–108 MV cm⁻¹ have extended the scope of optics¹⁴⁻¹⁶. CEP-stable waveforms have facilitated resonant control of low-energy elementary excitations¹⁸,¹⁹. Furthermore, these pulses may serve as ultrafast bias fields for extreme transport studies, because the photon energies lie far below the electronic interband resonances of bulk semiconductors¹⁷,¹⁹. Acting on the femtosecond scale, terahertz biasing is not limited by d.c. dielectric breakdown. Peak fields of 1 MV cm⁻¹ have shed new light on intervalley scattering or impact ionization¹¹. For similar amplitudes, an onset of ballistic transport through a fraction of the Brillouin zone of a bulk semiconductor has been reported¹. Yet, expectations are far ahead of experiments. A long-standing prediction by Bloch¹ and Zener² states that the quasi-momentum ħk of a crystal electron in an electric field E changes at a constant rate ħdk/dt = −eE, where ħ denotes the reduced Planck constant and e is the elementary charge. If E is large enough for k to reach the edge of the Brillouin zone before scattering takes place, Bragg reflection occurs and the electron traverses the Brillouin zone again¹⁴⁻¹⁸. This scenario causes charge oscillations in real and reciprocal space. When the electric bias competes with atomic potential gradients, extreme high-field phenomena such as interband ionization, Wannier–Stark localization and strong mixing of electronic bands may be expected. Although analogous phenomena have been studied in artificial structures²²⁻²⁷, the field-sensitive observation of coherent high-field transport in bulk solids has remained a challenge as the combination of ultrafast scattering¹ and the small size of the crystalline unit cell requires extremely strong biasing.

Figure 1 | Field-sensitive terahertz nonlinear optics. a, The waveform of the terahertz driving field (blue, solid curve) features a Gaussian envelope (black dashed curve) with an intensity full-width at half-maximum of 109 fs, which contains three optical cycles. The transient was recorded electro-optically in a GaSe sensor (thickness, 40 μm), with an 8 fs near-infrared gate pulse (centre wavelength, 0.84 μm). Inset: corresponding amplitude spectrum. b, Electro-optic trace of the waveform generated by the intense terahertz pulse of a in a GaSe single crystal (thickness, 220 μm; angle of incidence, θ = 70°), kept at room temperature. The data are shown as recorded with an AgGaS₂ detector (thickness, 100 μm), not corrected for the detector response, which leads to a temporal walk-off between the fundamental and the second harmonic. A superposition of the fundamental, second harmonic and optical rectification components is clearly seen. Insets: corresponding amplitude spectrum and experimental geometry, indicating the angle of incidence θ. The role of in-plane rotation of GaSe (angle ϕ) is investigated in Supplementary Fig. 5.

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Here, we use CEP-stable terahertz waveforms with amplitudes comparable to atomic fields to explore a qualitatively new regime of ultrafast all-coherent charge transport. The field drives interband polarization in a bulk semiconductor off-resonantly and accelerates the excited electron–hole pairs to perform dynamical Bloch oscillations, generating CEP-stable radiation throughout the frequency range between 0.1 and 675 THz. Quantum interference of multiple excitation channels makes the dynamics highly sensitive to the CEP of the terahertz waveform.

Intense phase-locked terahertz transients are generated by difference frequency mixing of two phase-correlated infrared pulse trains. Figure 1a shows the waveform detected electro-optically (centre frequency, 30 THz; peak field in air, $E_0 = 72$ MV cm$^{-1} = 0.7$ V Å$^{-1}$). The field is focused into a bulk crystal of the semiconductor gallium selenide (GaSe), which forms a perfect host for coherent high-field transport. As the widths of the valence and conduction bands are substantially smaller than the fundamental energy gap $E_g$, ultrafast carrier scattering by impact ionization is suppressed. The pulse emitted from the sample differs sharply from the incident wave. In the electro-optic signal, recorded with an AgGaS$_2$ sensor (Fig. 1b), a high-frequency component at the second harmonic and a low-frequency offset are superimposed on the fundamental wave. The inset to Fig. 1b depicts the corresponding amplitude spectrum. Emission between 0.1 and 10 THz originates from optical rectification, while the prominent peak at 60 THz stems from second-harmonic generation (see Chapter 5 in the Supplementary Information for phase-matching considerations). The shape of the electro-optic waveform remains constant for consecutive scans, underpinning the absolute phase stability of all frequency components.

Beyond second-order nonlinearities, ultrabroadband electro-optic traces also exhibit peaks at the third (90 THz) and fourth (120 THz) harmonics (Fig. 2a). Using a grating spectrometer with an InGaAs array detector, we record maxima at the sixth to ninth harmonics, with decreasing intensity. A monochromator and a cooled silicon charge-coupled device (CCD) monitor high-order harmonics (HH) with comparable intensities up to the 16th order (480 THz), which nearly coincides with the fundamental energy gap of GaSe at 476 THz. Interband photoluminescence attests to the presence of terahertz-induced electron–hole pairs. The existence of a plateau-like region is a hallmark of non-perturbative nonlinearities. HH generation (HHG) continues up to the 22nd order, despite strong interband absorption. The total spectrum of 23 harmonic orders (including optical rectification) covers 12.7 octaves from 0.1 to 675 THz (wavelength range, 3 mm to 440 nm), constituting a record bandwidth for tabletop terahertz sources. It is interesting to trace the HH intensity as a function of the terahertz amplitude, as shown, for example, for the 13th order in Fig. 2b. Although the intensity initially scales asymptotically as $I_{13} \propto E_0^{12}$ (dashed line in Fig. 2d), a slower increase of $I_{13} \propto E_0^{11}$ (dotted line in Fig. 2d) observed at the highest fields confirms the non-perturbative
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colour maps, for peak electric fields highlight the long-term stability of the spectral phase with a root within the HH comb, while repeated scans (Supplementary Fig. 2) monic. The fringe visibility of 81% proves excellent phase-coherence the interferogram of the 12th and the frequency-doubled sixth har-
monic. The nature of HHG. Other harmonic orders feature similar behaviour (Supplementary Fig. 7).

Importantly, the entire HH spectrum is phase-locked. The CEP stability of the low-frequency components has been established by electro-optic sampling (Fig. 1). Spectral interferometry of the HH comb with its own second harmonic allows us to test the CEP stability above electro-optically accessible frequencies. Stable interference between the harmonic wave of order 2n and the second harmonic of the nth harmonic is expected only if there is phase coherence between various orders. As an example, Fig. 2c represents the interferogram of the 12th and the frequency-doubled sixth harmonic. The fringe visibility of 81% proves excellent phase-coherence within the HH comb, while repeated scans (Supplementary Fig. 2) highlight the long-term stability of the spectral phase with a root

mean square (r.m.s.) jitter of only 0.4 µrad. To the best of our knowledge, these results mark the first direct verification of CEP locking of HH radiation and point towards all-coherent electronic dynamics as its microscopic origin.

HHG in atoms has been explained semiclassically by a three-step model of consecutive ionization, acceleration and recollision. In contrast, terahertz acceleration of electrons in a periodic lattice potential is dominated by Bragg reflection due to the quantum-mechanical wave nature of the electron. This dynamics is best described within a periodic band structure. First models of HHG in solids have captured the acceleration of pre-existing electrons in the conduction band, for example, by integrating Bloch’s acceleration theorem for a time-dependent external field. However, this approach does not explain the creation of an electronic population in the conduction band of an intrinsic semiconductor. For a consistent quantum description, we customize the theory of ref. 10, accounting for both off-resonant excitation of interband polarization and intraband acceleration of electronic wavepackets throughout the Brillouin zone (Supplementary Fig. 3). A realistic representation of terahertz-induced dynamical band mixing is obtained by including three valence and two conduction bands (Fig. 2d). This approach incorporates the broken inversion symmetry of GaSe, required for the generation of even harmonic orders. Evaluating this model for the parameters of bulk GaSe, we obtain the spectrum shown as a dashed curve in Fig. 2a. The theory describes the occurrence of even and odd harmonic orders and yields good agreement with the experimentally observed relative intensities.

Our calculations identify dynamical Bloch oscillations combined with coherent interband excitation as the physical origin of the HHG process. Figure 3 shows the computed dynamics of electron wavepackets for three different terahertz amplitudes. Electron-hole pairs are mainly created at negative field maxima (Fig. 3a,b) by coherent interband polarization. Simultaneously, each half cycle of the carrier wave drives the electron momenta from zero up to their maximal values and back to zero. In the central part of the medium- to high-intensity pulses (Fig. 3c,d), electrons are sufficiently accelerated to reach the Brillouin zone edge and undergo more than one complete Bloch cycle within one half-cycle of the terahertz wave, corresponding to a Bloch period of only a few femtoseconds. This dynamics leads to high-frequency radiation (Fig. 2a), which is coherently locked to the driving field (Fig. 2c). The highest harmonic orders are predominantly caused by the coherent intraband current, as confirmed by a switch-off analysis (Supplementary Fig. 4), a semiclassical estimate (Supplementary Fig. 9) and the dependence of HHG on crystal orientation (Supplementary Fig. 5).

Because the terahertz-induced coherent interband transitions involve several electronic bands, simultaneously, Bloch oscillations occur via a quantum interference of multiple excitation pathways (Fig. 2d). Accordingly, the instantaneous phase and amplitude of the exciting terahertz field sensitively control the wavepacket dynamics (Supplementary Fig. 3), and we expect HHG to depend on the CEP \( \varphi_{\text{CEP}} \) of the terahertz wave. Figure 4 summarizes the measured and computed CEP dependence of the HH spectrum. For \( \varphi_{\text{CEP}} = 0.1\pi \), the HH spectrum is approximately sinusoidally modulated with a period of 30 THz, up to the bandgap (Fig. 4a). Beyond 476 THz, the intensity decreases due to interband absorption. For its sign-flipped counterpart, that is, \( \varphi_{\text{CEP}} = 1.1\pi \), the shapes, the contrast and the magnitudes of the HH peaks change, corroborating the interference between different excitation paths. For a comprehensive picture, we recorded (Fig. 4b) and computed (Fig. 4c) intensity spectra while varying \( \varphi_{\text{CEP}} \) continuously from \( -\pi \) to \( 4\pi \). HHG below 400 THz is only slightly affected by the CEP. In contrast, the intensity maximum of the 16th harmonic shifts about a frequency of
multi-terahertz transients at the difference frequency (tuning range of centre frequency, 10–72 THz; pulse energy, up to 30 µJ). The CEP was controlled via the relative delay between the near-infrared generation pulses. This principle has been described in detail in ref. 15. A synchronized 8 fs near-infrared pulse (centre wavelength, 840 nm; pulse energy, 10 nJ) served as a gate for ultrabroadband electro-optic sampling. All experiments were performed at room temperature.

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Author contributions
O.S., M.H., F.L. and R.H. conceived the study. O.S., M.H., F.L., B.U., C.L. and R.H. carried out the experiment. U.H., D.G., T.M., M.K. and S.W.K. developed the quantum-mechanical model and carried out the computations. O.S., M.H., F.L., U.H., M.K., S.W.K and R.H. wrote the manuscript. All authors discussed the results.

Additional information
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Competing financial interests
The authors declare no competing financial interests.