Lightwave-driven quasiparticle collisions on a subcycle timescale

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Ever since Ernest Rutherford scattered α-particles from gold foils1, collision experiments have revealed insights into atoms, nuclei and elementary particles. In solids, many-body correlations lead to characteristic resonances—called quasiparticles—such as excitons, dropletons4, polarons and Cooper pairs. The structure and dynamics of quasiparticles are important because they define macroscopic phenomena such as Mott insulating states, spontaneous spin–charge-order, and high-temperature superconductivity5. However, the extremely short lifetimes of these entities6 make practical implementations of a suitable collider challenging. Here we exploit lightwave-driven charge transport7–10, the foundation of attosecond science11–13, to explore ultrafast quasiparticle collisions directly in the time domain: a femtosecond optical pulse creates excitonic electron–hole pairs in the layered dichalcogenide tungsten diselenide while a strong terahertz field accelerates and collides the electrons with the holes. The underlying dynamics of the wave packets, including collision, pair annihilation, quantum interference and dephasing, are detected as light emission in high-order spectral sidebands14–19 of the optical excitation. A full quantum theory explains our observations microscopically. This approach enables collision experiments with various complex quasiparticles and suggests a promising new way of generating sub-femtosecond pulses.

Following the principle of high-energy accelerators, their solid-state counterparts should prepare, accelerate and collide quasiparticles while detecting the outcomes. This can be realized only if all steps are accomplished faster than the ultrashort lifetime of the quasiparticle. Preparing the quantum states of quasiparticles by femtosecond laser is already well established20. Ultrafast acceleration, in turn, may harness the latest breakthroughs of strong-field control: the carrier wave of an intense laser pulse has been applied to ionize atoms or molecules and recollide their fragments when the sign of the light-field changes19. The excess energy of the collision is released as high-order harmonics of the laser frequency, which may emerge in attosecond bursts6, carrying the data to extract key information on the colliding species. Finally, we compare the process to HHG in the same material.

As a member of the class of transition metal dichalcogenides, WSe2 features strongly bound Wannier excitons21–23 and unique spin–valley coupling24. Even in its bulk form, WSe2 has been reported to exhibit an exciton binding energy of the order of 0.1 eV at its direct bandgap25, which implies that these quasiparticles are stable at room temperature. In a first experiment, a 100-fs excitation pulse (red waveform in Fig. 1a) centred at a photon energy of 1.621 eV (centre frequency, 392 THz) resonantly prepares coherent excitons at the direct band gap of WSe2 (thickness, 60 nm) located at the K and K’ points in momentum space. A co-propagating intense THz pulse (blue waveform in Fig. 1a) at its excitonic origin. Apart from the broadening of the sidebands due to photon emission by pair annihilation, which is detected as high-order sideband order, and even saturates at the highest field strengths (Fig. 1c, see also Extended Data Fig. 2 for the influence of the interband transition period T1 THz = 1/ν1 THz). Spectral components up to n = 2 are clearly visible, while the intensities of negative orders drop much faster with decreasing frequencies. The new frequency components feature the same polarization as the interband excitation pulse (Extended Data Fig. 1). In contrast to perturbative scaling (I HSG ∝ E2 peak), the sideband intensity grows linearly with E peak for intermediate peak fields, depending on the interband order, and even saturates at the highest field strengths (Fig. 1c, see also Extended Data Fig. 2 for the influence of the interband transition fluence). Finally, I HSG peaks if the interband excitation is resonant with the excitonic absorption peak (Fig. 1d), underpinning its excitonic origin. Apart from the broadening of the sidebands due to the ultrashort duration of E1 THz, these findings agree well with previous time-integrated studies16.

To directly resolve the underlying subcycle quantum dynamics, we now prepare the coherent excitons within 10 fs—much faster than a single oscillation period T = 1/ν1 THz of the THz wave. Figure 2a depicts the spectral shape of the observed sideband intensity I HSG as a function of the delay between the excitation pulse and the peak of the THz driving field, t ex. Electro-optic detection allows us to determine the complete waveform E1 THz on the same absolute timescale (Fig. 2a, blue curve). The spectrogram in Fig. 2a follows a nearly Gaussian temporal envelope, resembling the THz envelope. Interestingly, I HSG is strongly modulated along the time axis with a period of 7T/2; that is, there is a subcycle criterion for ‘good’ and ‘bad’ preparation times t ex.
For a quantitative timing analysis, Fig. 2b compares $E_{\text{THz}}$ (blue curve) with the spectrally integrated intensity $I_{\text{HSG}}$ (red shaded curve). The strongest THz half-cycle occurs at a delay $\delta_{\text{global}} \approx T$ with respect to the most-intense sideband peak. This retardation rules out instantaneous optical nonlinearities as the microscopic origin of HSG. A detailed comparison of $E_{\text{THz}}$ and $I_{\text{HSG}}$ also reveals a subcycle delay $\delta_{\text{ec}}$ of the observed sideband bursts relative to the THz crests (see inset in Fig. 2b). For subsequent half-cycles of the driving field, $\delta_{\text{ec}}$ initially increases, reaches a maximum value of approximately $T/8$ at the centre of the THz pulse and slightly decreases afterwards (Fig. 2d).

To link these signatures of $I_{\text{HSG}}$ with the microscopic quasiparticle motion, we analyse the experiment using a full quantum mechanical model based on the semiconductor Bloch equations (see Methods section ‘Microscopic model of HSG’). For a 100-fs preparation pulse, the experimentally observed HSG spectrum is well reproduced by our calculations (Fig. 1b, black dashed curve), showing a similar modulation and intensity roll-off on the high- and low-frequency sides. Using a 10-fs preparation pulse, the calculations quantitatively reproduce the temporal characteristics of $I_{\text{HSG}}$ (Fig. 2c), including even delicate features such as $\delta_{\text{ec}}$ (Fig. 2d).

Figure 3 visualizes the underlying microscopic quasiparticle dynamics for two characteristic preparation times $t_{\text{ex}}$, corresponding to minimum ($t_{\text{ex}} = -4\text{ fs}$, Fig. 3c, e, g) and maximum ($t_{\text{ex}} = 7\text{ fs}$, Fig. 3d, f, h) HSG. The two quasiparticle-collider sequences are schematically illustrated by Feynman diagrams in Fig. 3a, b. For $t_{\text{ex}} = -4\text{ fs}$, the coherent excitons are prepared shortly before a THz field crest (see vertical dashed lines in Figs 2c and 3c). Immediately after optical preparation, the average momenta of the electron (Fig. 3e, solid curve) and the hole (dashed curve) are located in the vicinity of the K and K’ points of the Brillouin zone (defined as $k = 0$). The strong THz field accelerates the distribution function of the electron ($f_{\text{e}}^{\prime}$; Fig. 3e, false-colour plot) and the hole ($f_{\text{h}}^{\prime}$; not shown) away from $k = 0$, driving the electron and hole wave packets apart. A qualitatively different situation occurs for $t_{\text{ex}} = 7\text{ fs}$ (Fig. 3d, f, h), for which the coherent excitons are prepared shortly before a zero crossing of the THz field (vertical dashed lines in Figs 2c and 3d). Now the mean momenta change sign; that is, the initially separating electron and hole are set on a recollision path (Fig. 3f).

Particle preparation of typical accelerators is spatially precise and collisions are monitored in real space. Our concept follows the relative motion of quasiparticles, and the preparation is precise in terms of the relative coordinates of the quasiparticles because the optical pulse creates them in a single, spatially correlated excitonic state described by a pair correlation function (see Methods section ‘Coherent electron–hole pair–correlation function’). Figure 3g, h displays its coherent part $g_{\text{coh}}(r)$, which defines how electrons are coherently coupled to holes as a function of their relative distance $r$ (reciprocal space representation in Extended Data Fig. 3). In both panels, $g_{\text{coh}}(r)$ is maximum close to the peak of the optical pulse before it decays by ultrafast dephasing (dephasing time $T_2 < T/2$). Figure 3c, d shows the corresponding mean electron–hole separation ($\bar{r}$). Suitable $t_{\text{ex}}$ (Fig. 3d) allows the THz field (Fig. 3c, d, blue curve) to separate and collide the electron–hole pairs at high energy. Owing to this ballistic motion, maximum HSG occurs for preparation times that are offset with respect to the field crests, causing the distinct delay $\delta_{\text{ec}}$ observed in Fig. 2d. The precise temporal structure seen in the experiment can be reproduced only if...
excitation-induced dephasing\textsuperscript{37} is taken into account (see Methods section ‘Effect of excitation-induced dephasing on HSG’ and Extended Data Figs 4–6). By contrast, for ‘bad’ timing $t_{\text{ex}}$, the electron and the hole are monotonically driven apart (Fig. 3c), suppressing collisions (see Fig. 3a). The situation is analogous to a cyclotron, in which electrons are effectively accelerated only if they are injected at the right phase of the alternating accelerator field.

Whereas the average intraband dynamics ($r$) resembles semiclassical trajectories (see Fig. 3c, d), the connection between quasiparticle collision and HSG requires a full quantum theory. Similarly to electron–positron pairs in vacuum, colliding electron–hole pairs in a solid are subject to pair annihilation, during which a high-energy photon may be emitted, leading to sideband signals; this microscopic annihilation dynamics manifests itself in $g_{\text{coh}}(r)$ (Fig. 3h). After substantial spreading of $g_{\text{coh}}(r)$, electron–hole collision is prompted by an abrupt termination of the correlation function due to pair annihilation. Subsequently, interference patterns occur among the residual fragments that survive the collision. This unique characteristic of the quasiparticle collider becomes particularly clear when we compare the dynamics with HHG in the same sample, which is achieved by applying the strong THz field without optical preparation of excitons. A typical high-harmonic spectrum exhibiting odd orders up to $n' = 47$ is depicted in Extended Data Fig. 7. The intensity of HHG is smaller by approximately one order of magnitude than HSG for frequencies below 650 THz. Owing to the non-perturbative, non-resonant excitation of Bloch electrons with simultaneous acceleration by the strong-field bias\textsuperscript{20,21,24,28}, the initial carrier distribution is smeared out throughout the entire Brillouin zone. Furthermore, coherent interband polarization can emit photons at any point in momentum space (see inset of Fig. 4b). This situation manifests itself by a critical dependence of the intensity and the shape of high-order harmonics on $\varphi$ (Fig. 4b), reflecting the six-fold symmetry of WSe\textsubscript{2} in the layer plane.

In conclusion, we demonstrate the realization of quasiparticle collisions in the time domain. We excite, accelerate and collide coherent electron–hole pairs in WSe\textsubscript{2} resulting in ultrabroad sideband emission. On a subcycle scale, the recorded sideband intensity (red) peaks at a distinct time delay $\delta_{\text{ex}}$ after the nearest extrema of the driving waveform (see close-up in b).

Well-defined initial momenta and relative spatial coordinates of the electrons and holes are prerequisites for controlled collisions. By contrast, the direction of acceleration in the WSe\textsubscript{2} crystal is less relevant, as seen in Fig. 4: the high-order sideband spectra show virtually no dependence on the azimuthal in-plane angle $\varphi$ of the THz polarization. This behaviour is compatible with our theoretical expectation that coherent electron–hole wave packets in HSG remain rather close to $k = 0$ (Fig. 3f). Both Coulomb attraction between electrons and holes and fast dephasing keep the coherent electron–hole excitations within a region in which the effective mass is approximately isotropic (see Fig. 1a). Additionally, HSG probes the interband polarization of colliding electrons and holes only at spatial overlap\textsuperscript{37} (inset of Fig. 4a).

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Quantum simulation of subcycle electron-hole collisions underlying HSG. 

- **Figure 3**
  - **a, b.** Schematic Feynman diagrams depicting electron–hole (e–h) pair creation by a near-infrared photon (hνopt) and acceleration by EThz, only for ‘good’ excitation times (b). The acceleration leads to a collision and annihilation of the electron–hole pair, thereby emitting a sideband photon hνHSG. **c, d.** Trajectories (red, the intensity of the line represents the density of coherent excitons) tracing the real-time evolution (time t) of the mean electron–hole separation (weighted average of gcoh(r)). For characteristic delays t ≈ 4 fs, corresponding to minimum (c) and maximum (d) HSG emission (compare with Fig. 2c). Vertical black dotted lines highlight t = 0, which marks the peak of the excitation pulse. Although electrons and holes are initially separated, they rapidly recollide (zero excursion marked by the black horizontal line); time of recollision is highlighted by the black arrow in d upon reversal of the driving field (blue curves). Induced by a strong multi-THz excitation of a bound electron–hole pair (grey), the carriers (red and blue wave packets) are accelerated and recollided by the strong light field (blue) to recombine at r = 0, giving rise to high-order sideband emission.

- **Figure 4**
  - **a, b.** Inset, schematic of high-order harmonic generation: a polarization between the conduction and valence bands (grey) is induced by a strong multi-THz field (blue) and simultaneously accelerated within the bands (red and blue spheres, curved red and blue arrow). During this process, the coherent interband polarization is continuously modified (red vertical arrows). The high-harmonic spectra reflect the six-fold symmetry of WSe2. **Inset,** experimental comparison of high-order sideband and high-order harmonic generation: a, b, Spectrally resolved high-order sideband intensity (colour scales) are normalized to the maximum HSG signal. Both intensity (colour) scales are normalized to the maximum HSG signal. Whereas high-order sidebands show virtually no dependence on ϕ, the high-harmonic spectra reflect the six-fold symmetry of WSe2. Inset, real-space visualization of THz-driven electron–hole recollisions: upon excitation of a bound electron–hole pair (grey), the carriers (red and blue wave packets) are accelerated and recollided by the strong light field (blue) to recombine at r = 0, giving rise to high-order sideband emission.
density of quasiparticles in solids helps to generate relatively intense, all-coherent, ultrabroadband radiation, which may pave the way to new sub-femtosecond light sources; furthermore, the subcycle modulation of HSG may facilitate information processing at optical-clock rates. Large-scale colliders have enabled the detailed study of elementary particles. Likewise, we anticipate that THz-driven, real-time collisions—operating on much smaller, widely tunable, length and energy scales—will be key to studying complex quasiparticles in numerous applications of modern materials science; examples include excitons, trions, biexcitons or dropletions in quasi-two-dimensional atomic monolayer systems, Dirac-like quasiparticles in graphene and topological insulators, or polarons in materials featuring strong electron–phonon coupling. This scheme may also be extended to optically excited quasiparticles in unconventional superconductors, and could help to resolve some of the outstanding questions in condensed matter research.

Online Content Methods, along with any additional Data display items and Source Data, are available in the online version of the paper; references unique to these sections appear only in the online paper.

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Author Information Reprints and permissions information is available at www.nature.com/reprints. The authors declare no competing financial interests. Readers are welcome to comment on the online version of the paper. Correspondence and requests for materials should be addressed to R.H. (rupert.huber@physik.uni-regensburg.de) or M.K. (mackillo.kira@physik.uni-marburg.de).
**METHODS**

**Experimental set-up.** Intense, phase-locked waveforms in the far- to mid-infrared spectral region (multi-terahertz range) are generated by difference frequency mixing of two spectrally detuned, near-infrared pulse trains from a dual optical parametric amplifier pumped by a titanium-sapphire laser amplifier\(^{11,12,14}\). The few-cycle transients feature peak fields of up to approximately 1 V Å\(^{-1}\) (see Fig. 1d). For the time-resolved measurements, the bandwidth of the excitation pulse is adjusted in the Fourier plane of a prism compressor, which enables the required pulse duration of 10fs. Spectral components of the excitation pulse above 400 THz are filtered out to avoid the resonant generation of a high density of unbound electron–hole pairs. All experiments were performed at room temperature and in ambient air. No sample degradation was observed during the whole course of experiments.

The spot sizes of the excitation pulse and the THz pulse at the sample are 22 μm and 85 μm, respectively (intensity full-width at half-maximum). The generated high-order bandwidth intensity \(I_{100}\) and the high-order harmonic intensity \(I_{101}\) are recorded with a spectrograph featuring a cooled silicon CCD camera. All spectra are corrected for the grating efficiency and the quantum efficiency of the detector.

The pulse energy of the narrowband excitation amounts to approximately 0.1 nJ. The few-cycle transients feature peak fields of up to approximately 1 V Å\(^{-1}\) (see Fig. 1d). For the time-resolved measurements, the bandwidth of the excitation pulse is adjusted in the Fourier plane of a prism compressor, which enables the required pulse duration of 10fs. Spectral components of the excitation pulse above 400 THz are filtered out to avoid the resonant generation of a high density of unbound electron–hole pairs. All experiments were performed at room temperature and in ambient air. No sample degradation was observed during the whole course of experiments.

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can contain different aspects of the collisions. As in conventional colliders, the scattering-generated radiation produces a wealth of information about the (quasi) particles. Here, we study the HSG intensity $I_{\text{HSG},\nu}$ as a function of the HSG frequency $\nu_{\text{HSG}}$, the field strength $E_0$, the duration $\tau_s$ and the central frequency $\nu_0$ of the optical preparation pulse ($\lambda = \text{opt}$) and the THz waveform ($\lambda = \text{THz}$), and their mutual delay $\Delta_{\text{opt-THz}}$ generating at least eight-dimensional spaces $I_{\text{HSG}(\nu_{\text{HSG}}, \nu_0, \lambda_{\text{opt}}, \lambda_{\text{THz}}, T_{\text{THz}}, t_{\text{opt}}, t_{\text{THz}}, \nu_{\text{HSG}}, \nu_{\text{THz}}, \Delta_{\text{opt-THz}})$.

As is typical for colliders, such a wealth of data reveals details of colliding quasiparticles only through a dedicated identification process. For example, Fig. 3 demonstrates how changing $t_{\text{opt}}$ yields a direct detection of the electron–hole recollision. We will next illustrate a few additional possibilities to extract excitation-related details from the HSG scattering data.

As shown in Extended Data Fig. 4, broadening of sideband resonances is directly related to excitation decay rates, which depend on the excitation parameters. This effect is often referred to as EID44,45, which can be controlled, for example, by changing $E_{\text{opt}}$ (ref. 34). We have also confirmed that the excitation binding energy $E_B$ strongly influences the overall strength of the HSG signal. For example, a tenfold increase of $E_B$ amplifies $I_{\text{HSG}}$ by roughly the same amount as a result of Coulomb enhancement of the collisional cross-section. Hence, a variety of material- and excitation-dependent exciton features may be deduced by quantitatively analysing the strength and width of $I_{\text{HSG}}$ resonances.

By changing the optical pump frequency $\nu_{\text{opt}}$, one can deduce differential changes in HSG spectra that sensitively monitor material details. We propose measuring a 2D $I_{\text{HSG}(\nu_{\text{HSG}}, \nu_{\text{opt}})}$ to set to follow spectral changes at the second-order sideband to define a normalized spectrum

$$I(\nu, \nu_{\text{opt}}) = \frac{I_{\text{HSG}(\nu_{\text{HSG}} - \nu_{\text{opt}}, \nu_{\text{opt}})}}{I_{\text{SB}}}$$

in which $\nu_{\text{opt}} = \nu_{\text{opt}} - \nu_{\text{opt}}$ is the frequency scale centred around $\nu_{\text{opt}}$ and $I_{\text{SB}} = I_{\text{HSG}(\nu_{\text{HSG}}, \nu_{\text{opt}})} + 2hI_{\text{THz}}$, the peak intensity of the second-order sideband. This normalized presentation spectra that do not depend on the absolute intensity scale—a quantity that is difficult to determine experimentally for a wide range of frequencies $\nu_{\text{opt}}$. From the difference of two spectra measured with pump frequencies $\nu_{\text{opt}} = \pm \Delta_{\text{opt}}/2$, we deduce a finite differential

$$\Delta I(\nu, \nu_{\text{opt}}) = \frac{1}{\Delta_{\text{opt}}}(I(\nu, \nu_{\text{opt}}) - \Delta_{\text{opt}}/2) - I(\nu, \nu_{\text{opt}} + \Delta_{\text{opt}}/2)$$

which approaches the derivative $\frac{I}{\Delta \nu_{\text{opt}}}$ in the limit $\Delta_{\text{opt}} \to 0$.

Extended Data Fig. 8a, b shows computed $\Delta I(\nu, \nu_{\text{opt}})$ for two 1s–1s excitation binding energies, $E_B = 60$ meV and $E_B = 600$ meV, respectively, when $h\nu_{\text{opt}}$ is expressed in terms of the detuning $\Delta_{\text{opt}}$ with respect to the 1s–1s excitation energy $h\nu_{\text{opt}}$, as $h\nu_{\text{opt}} = h\nu_{\text{opt}} + \Delta_{\text{opt}}$. These contours produce spectral information that differs greatly for the two excitation binding energies, demonstrating that $\Delta I(\nu, \nu_{\text{opt}})$ exposes material-dependent properties. For $E_B = 60$ meV, $\Delta I(\nu, \nu_{\text{opt}})$ exhibits negative changes for $\Delta_{\text{opt}} < 0$, implying that $I_{\text{HSG}}$ decreases for negative detuning. Because $\Delta_{\text{opt}} > 0$ yields mainly positive $\Delta I(\nu, \nu_{\text{opt}})$, a clear dispersive shape is observed around the resonant excitation $\nu_{\text{opt}} = 0$. For $E_B = 600$ meV, a positive island appears within the $\Delta_{\text{opt}} < 0$ region, which implies that $I_{\text{HSG}}$ Can be enhanced also for negative $\Delta_{\text{opt}}$, in contrast to the $E_B = 60$ meV case. Additionally, $\Delta_{\text{opt}} > 0$ produces a new feature because the positive islands split into a double-peak structure for $E_B = 600$ meV.

To analyse the quantitative dependence of $\Delta I(\nu, \nu_{\text{opt}})$ on $E_B$, Extended Data Fig. 8c, d shows slices of $\Delta I(\nu, \nu_{\text{opt}})$ at fixed values of $h\nu_{\nu_{\text{opt}}} = \pm 16$ meV, respectively, for three different binding energies $E_B = 60$ meV (black curve), $240$ meV (red curve) and $600$ meV (blue curve). The slice at $-16$ meV shows only one positive peak for $\Delta_{\text{opt}} > 0$ and remains negative at $\Delta_{\text{opt}} < 0$ for $E_B = 60$ meV (black curve). Increasing the excitation binding energy gradually creates a new maximum within the region of negative detuning. This transition from a single- to a double-peak structure can be exploited to assign the excitation binding energy as shown below. Monitoring the slices at $h\nu_{\text{opt}} = -16$ meV reveals a smooth, narrow, single-peak structured close to $\Delta_{\text{opt}} = 0$ for $E_B = 60$ meV (black curve). By contrast, higher binding energies produce a more structured shape.

To construct differential spectra $\Delta I(\nu, \nu_{\text{opt}})$ experimentally, we measure HSG spectra for narrowband pump pulses at different frequencies $\nu_{\text{opt}}$ (see Fig. 1b, d) while varying $\nu_{\text{opt}}$. Extended Data Fig. 9c shows the resulting intensity map $I_{\text{HSG}(\nu_{\text{HSG}}, \nu_{\text{opt}})}$ for resonant excitation $h\nu_{\text{opt}} = h\nu_{\text{opt}}$, the corresponding computation is shown in Extended Data Fig. 9e. Both experiment and theory show the same spectral structure, including $\nu_{\text{opt}}$-dependent changes in the intensity and shape. A similar level of agreement is realised also for non-resonant excitations (not shown), paving the way to thorough theory–experiment comparison of the differential spectra.
On the basis of Extended Data Fig. 8, we expect the strongest dependence of $\Delta I(\nu, \nu_{\text{opt}})$ on $E_B$ for a finite detuning $\Delta_{\text{opt}}$ from the $1s$-exciton resonance. We therefore compare experimental traces $\Delta I(\nu, \nu_{\text{opt}})$ at $E_B = -20\text{fs}$ for three different detunings $\Delta_{\text{opt}} = -75\text{meV}$ (shaded area), $-29\text{meV}$ (black curve) and $27\text{meV}$ (red curve) in Extended Data Fig. 9a. A negative $\Delta_{\text{opt}}$ produces a dispersive line shape to $\Delta I(\nu, \nu_{\text{opt}})$, that is, a dip followed by a zero crossing and a peak, which implies a shift of the underlying HSG intensity with a change in $\nu_{\text{opt}}$. For positive $\Delta_{\text{opt}}$, a double-peaked structure indicates a broadening of the HSG peaks. For a binding energy of $E_B = 60\text{meV}$, these line shapes are well reproduced by the theoretical computations using the same detuning $\Delta_{\text{opt}}$ and excitation conditions as in the experiment, as shown in Extended Data Fig. 9b.

Extended Data Fig. 9d, f shows the corresponding calculated differentials $\Delta I(\nu, \nu_{\text{opt}})$ when the exciton binding energy is increased to $E_B = 240\text{meV}$ and $E_B = 600\text{meV}$, respectively. Whereas $\Delta_{\text{opt}} = -29\text{meV}$ (black curve) and $\Delta_{\text{opt}} = 27\text{meV}$ (red curve) produce a similar shape as for $E_B = 60\text{meV}$, the peak amplitude of the differential curve for $\Delta_{\text{opt}} = -75\text{meV}$ (shaded area) greatly decreases for an intermediate binding energy of $E_B = 240\text{meV}$ and turns into a negative dip for the largest binding energy. This structural change in $\Delta I(\nu, \nu_{\text{opt}})$ can be used to deduce the exciton binding energy. We find that the experimental data are consistent with $E_B = 60\pm 20\text{meV}$, which agrees well with values reported in literature.30,46,47

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Extended Data Figure 1 | Polarization of high-order sidebands. a, b, False-colour plot of the spectral intensity of high-order sidebands (generated under resonant, spectrally narrow, optical excitation) of orders two to eight as a function of their polarization for horizontal (a) and vertical (b) polarization of the interband excitation pulse. The polarization angle is defined such that $0^\circ$ corresponds to a horizontally polarized excitation. c, Spectrally integrated sideband intensity as a function of the polarization angle.
Extended Data Figure 2 | Pump-fluence dependence of time-resolved high-order sideband generation. a, The measured high-order sideband intensity $I_{HSG}$ (spectrally and temporally integrated, red spheres) scales linearly with the pump fluence $\Phi$, as indicated by a guide to the eye (black dashed line). b, The modulation depth of the spectrally integrated temporal trace of $I_{HSG}$ (red spheres, see Fig. 2b) decreases with increasing pump fluence, closely following a fit (black curve) proportional to the inverse square-root of the pump fluence. c, By contrast, the relative subcycle delay $\delta_{sc}\tau^{-1}$ does not substantially change with increasing pump fluence. Red spheres represent the average of $\delta_{sc}\tau^{-1}$ over the nine dominant, consecutive, high-order sideband peaks (see Fig. 2d) for different pump fluences; error bars indicate their standard deviation. The black dashed line marks the mean value of the displayed data points.
Extended Data Figure 3 | Coherent excitonic polarization dynamics in k space. a, Computed high-order sideband intensity $I_{\text{HSG}}$ (red shaded area) and driving waveform (blue). Vertical black dotted lines highlight characteristic delay times $t_{\text{ex}}$ at extrema of $I_{\text{HSG}}$. b–d, Coherent interband polarization $|p|^2$ (colour scale) as a function of time $t$ and reciprocal space coordinate $k$ (in units of the wave vector $k_{\text{BZ}}$ limiting the first Brillouin zone) for distinct delay times $t_{\text{ex}}$ according to maximum (b, $t_{\text{ex}} = 7$ fs; d, $t_{\text{ex}} = 28$ fs) and minimum (c, $t_{\text{ex}} = 18$ fs) HSG emission. Horizontal white lines mark $k = 0$. The driving field is depicted as grey curves and the recollision times (see Fig. 3d) are highlighted by black arrows.
Extended Data Figure 4 | Influence of dephasing on high-order sideband generation. a, b, The measured sideband spectrum (shaded area) is compared with computations using constant dephasing times of $T_2 = 3.2$ fs (a, red curve), $T_2 = 4$ fs (b, blue curve) and a momentum-dependent dephasing model as presented in Fig. 1 (black curves; EID, excitation-induced dephasing). The sideband orders $n$ are indicated above the relevant peaks. All spectra are normalized to the sideband peak corresponding to $n = 2$. The inset in a depicts the corresponding dephasing times $T_2(k)$ as a function of the wave vector $k$: the red (blue) horizontal line indicates a constant decay level $T_2 = 3.2$ fs ($T_2 = 4$ fs).
Extended Data Figure 5 | Field scaling of time-resolved high-order sideband generation. a, Scaling of the modulation depth (see discussion in Methods section ‘Influence of excitation fluence’) of temporal traces of high-order sideband intensity with the driving peak field strength. An increase in the modulation depth suggests a faster dephasing $T_2$. b, Subcycle delay $\delta_{sc}$ in units of the driving period $T$ averaged over the eight most-dominant sideband peaks as a function of the peak field. The error bars represent the standard deviation of the eight peaks and the dashed line depicts a linear fit to the data points.
Extended Data Figure 6 | Influence of excitation-induced dephasing. 

a, Terahertz waveform used in the computations. 

b, c, Mean electron excursion $\langle \Delta r \rangle_k$ calculated with a constant dephasing $T_2 = 12$ fs (b) and excitation-induced dephasing (c) for a delay of $t_{ex} = 7$ fs (see vertical dotted lines in a). The shaded areas indicate the intensity envelopes of the excitation pulse with a full-width at half-maximum of 10 fs (yellow) and 100 fs (grey). The corresponding mean momenta $\langle \Delta p \rangle_k$ are shown as red and black curves, respectively. The red dashed curves show $\langle \Delta p \rangle_k$ after the electron–hole collision. The dashed horizontal lines in c mark the positions in $k$ space where the scattering time $T_2(k)$ switches from slow to fast dephasing.
Extended Data Figure 7 | High-order harmonic generation in tungsten diselenide. An intensity spectrum (blue shaded area) of THz-driven high-order harmonic generation in 60-nm-thick WSe₂ shows distinct peaks at odd orders \( n' = 13 \) to \( n' = 47 \) (indicated by numerals) of the fundamental frequency \( \nu_{\text{THz}} = 22.3 \) THz (peak electric field in air, 21 MV cm\(^{-1}\); \( \varphi = 90^\circ \)). An intensity spectrum for \( \varphi = 120^\circ \) is also shown as a black curve. The spectral intensity has been corrected for the grating efficiency and for the quantum efficiency of the spectrograph used.
Extended Data Figure 8 | Differential sideband spectroscopy. 

a, b, Contours of differential spectra $\Delta I(\nu, \nu_{opt})$ (colour scale) as a function of $h(\nu - \nu_{2SB})$ and detuning $\Delta_{opt}$ computed for an excitonic binding energy of $E_B = 60$ meV (a) and $E_B = 600$ meV (b). The black and grey vertical lines mark the positions of the slices shown in c and d, respectively. 

c, d, Snapshots of $\Delta I(\nu, \nu_{opt})$ at fixed values of $|h(\nu - \nu_{2SB})| = 16$ meV below (c) and above (d) the second-order sideband peak for three different binding energies $E_B = 60$ meV (black curves), $E_B = 240$ meV (red curves) and $E_B = 600$ meV (blue curves).
**Extended Data Figure 9 | Quantitative analysis of the binding energy.**

**a.** Measured differential spectra $\Delta I(\nu, \nu_{\text{opt}})$ for three different detunings $\Delta_{\text{opt}} = -75$ meV (shaded area), $\Delta_{\text{opt}} = -29$ meV (black curve) and $\Delta_{\text{opt}} = 27$ meV (red curve) as functions of $\nu$, centred at the position of the second sideband $\nu_{2\text{SB}}$.

**b, d, f.** Computed differential spectra $\Delta I(\nu, \nu_{\text{opt}})$ for binding energies of $E_B = 60$ meV (b), $E_B = 240$ meV (d) and $E_B = 600$ meV (f) and detunings $\Delta_{\text{opt}}$ as in the experiment shown in a. **c, e.** Measured (c) and calculated (e) HSG intensities (colour scale) for $\Delta_{\text{opt}} = 0$ as functions of $\nu_{\text{HSG}}$ and delay $t_{\text{ex}}$. 

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