

Supplementary

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Contents

1	Experimental Setup	3
2	α- Quartz structure, symmetries and DFT calculations	4
3	Interband mechanism in α- Quartz HHG	7
4	Semi classical trajectory analysis	11
4.1	Single-color linearly-polarized light	14
4.2	Single-color elliptically-polarized light	15
4.3	Two-color cross-polarized light	17
5	HHG Spectroscopy in broken inversion crystals- Quantum analysis	20
5.1	Interband emission	20
5.2	Saddle point approximation	22
5.3	Discrete Berry phase	24
6	α-quartz HHG spectroscopy	27
6.1	Linearly Polarized Field	27
6.2	Elliptically Polarized Field	29
6.2.1	Reconstruction of the Berry Phase	31
6.2.2	Theoretical Berry phase calculation	32

6.3	Two-color Field	33
6.3.1	Theoretical study	37

1 Experimental Setup

The experiment was performed using a near-infrared laser source (Light Conversion TOPAS-HE) having a center wavelength of $\sim 1200\text{nm}$, at 1kHz repetition rate and 50fs pulse duration. Figure 1 schematically describes the experimental set up for

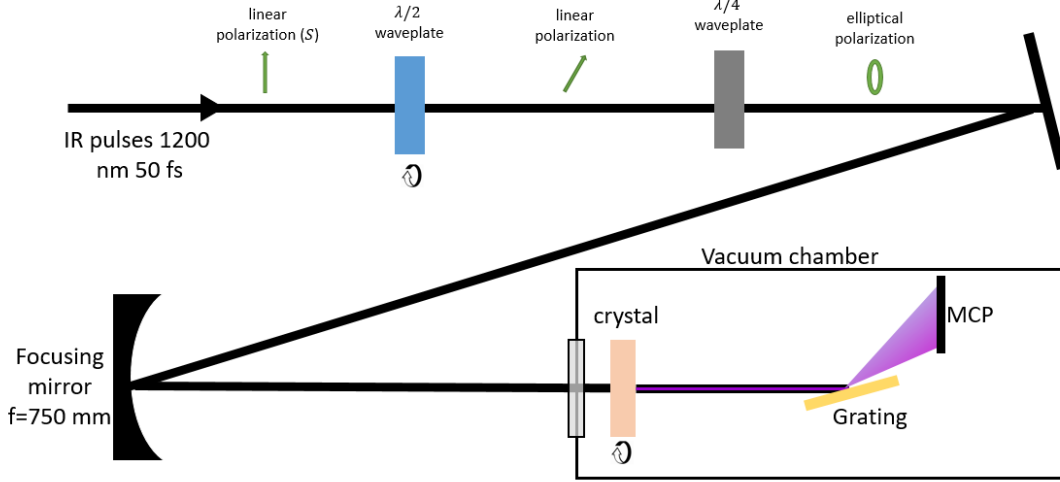


Figure 1

Schematic description of HHG spectroscopy setup, driven by an elliptically-polarized driving field.

HHG spectroscopy measurements, driven by an elliptically polarized light. The ellipticity of the laser field was controlled with a motorized half waveplate, followed by a quarter waveplate. This combination allows us to scan the ellipticity degree of the light, while keeping the main axis of the ellipse fixed. The intensity of the laser field is approximately $\sim 5 \cdot 10^{13} \frac{\text{W}}{\text{cm}^2}$. The beam was focused into a free-standing single crystal of c-cut ($< 100 >$) $20\mu\text{m}$ thick α -quartz sample (SiO_2) placed at the imaging plane of an XUV spectrometer. The orientation of the crystal axes with respect to the laser polarization was controlled by a rotation motor.

Figure 2 schematically describes the second part of the experiment, two-color HHG spectroscopy. A weak second harmonic (SH) field was generated using a $100\mu\text{m}$ type-I phase barium borate (BBO) (BaB_2O_4) crystal, orthogonally polarized

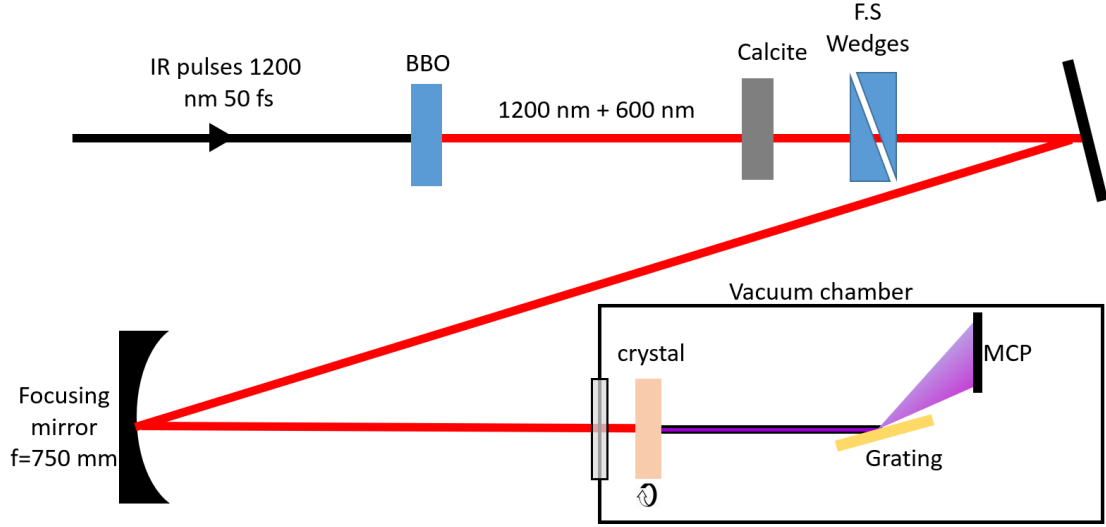


Figure 2: Two-color HHG spectroscopy setup

to the fundamental field's polarization. The intensity of the fundamental field is approximately $\sim 5 \cdot 10^{13} \frac{W}{cm^2}$, while the intensity of the SH field was less than 5% the fundamental intensity. A birefringent crystal (calcite) was used to compensate for group-velocity dispersion between the two fields, and their relative sub-cycle delay was controlled using a pair of fused silica wedges.

2 α - Quartz structure, symmetries and DFT calculations

α -quartz has a threefold rotational (C_3) symmetry around the c-axis, and a twofold symmetry (C_2) around the a-axis. Its space group is denoted as $P3_221$. For a laser field propagating in parallel to the c-axis, the HHG interband interaction can be described within the field's polarization plane, the c-plane. In the two-dimensional projection of quartz onto this plane, the C_2 symmetry of the crystal is equivalent to a mirror symmetry, as can be seen in the illustration in figure 3a. In addition, the atomic structure in the c-plane can be split into three identical patterns, repeated

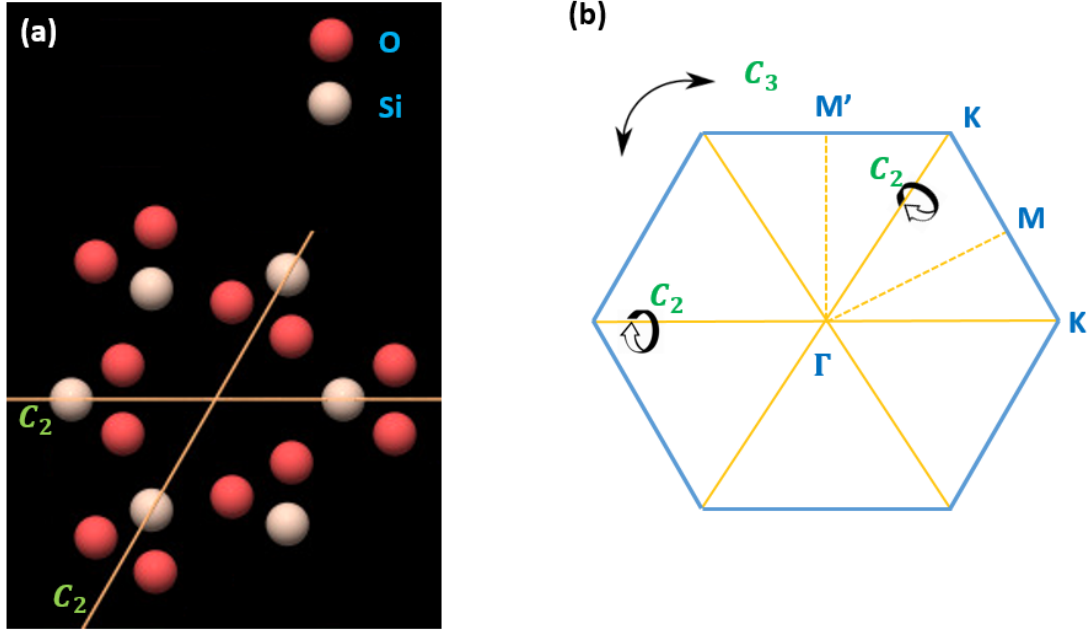


Figure 3: (a) An α -quartz projection of atoms onto the c -plane, demonstrating C_3 in-plane symmetry, and C_2 out-plane symmetry. The golden lines represent C_2 axes. (b) The first Brillouin zone of quartz, displaying the C_2 and C_3 symmetries in the reciprocal space. The solid golden lines correspond to the $\Gamma - K$ direction and the dashed lines show correspond to $\Gamma - M$ and $\Gamma - M'$.

every 120° , thus demonstrating C_3 symmetry. Additionally, the lines connecting two S_i atoms through the center correspond to the C_2 axes. To describe the dynamics of the crystal's electronic eigenmodes, it is useful to examine these symmetries in the reciprocal crystal momentum space (k -space). Accordingly, figure 3b depicts the first Brillouin zone (BZ) of quartz, highlighting the main symmetry axes, $\Gamma - K$ and $\Gamma - M$, corresponding to the 0° and 30° rotational directions in the main text, respectively. Each $\Gamma - K$ line coincides with a C_2 symmetry axis. The BZ is symmetric with respect to a 120° rotation, thus respecting the C_3 symmetry as well.

The electronic structures were extracted by first principle calculations, performed in the frame work of density-functional theory by the Vienna Ab initio Simulation

Package (VASP) [1, 2]. The electron exchange-correlation effect is described by the Perdew-Burke-Ernzerhof (PBE) formulation, under the generalized gradient approximation (GGA)[3]. The projector augmented wave (PAW) pseudopotential [4, 5] was used to describe the interaction between electrons and nuclei. An energy cut-off as 520 eV was employed for the plane wave basis. The $10 \times 10 \times 10$ Γ -centered Monkhorst-Pack grids were employed to sample the Brillouin zone for the relaxation of all structures. The Wannier function-based tight-binding Hamiltonian of alpha-quartz is obtained by generating the maximally localized Wannier functions from the Wannier90 software [6], which is interfaced to VASP. The electronic structures and Berry phase related properties are calculated from the tight-binding-type Hamiltonian.

The band structure of quartz is displayed in figure 4a. The highest valence band (band 12, green curve around 0eV) is separated from the lowest conduction band (band 13, light blue curve) by a large band gap, having a minimal value of $\sim 10.4\text{eV}$ around the Γ point. By closely examining the conduction bands, one can notice a few distinctive features of the lowest one. Compared with the other conduction bands, it spans over the largest energy range across the BZ, while retaining the largest band gap from all other conduction bands. These features allow us to study this band's k -space intrinsic properties as well as their mapping to energy via the emitted HHG.

Finally, we describe the intraband Berry curvature, defined as:

$$\boldsymbol{\Omega}_n(\mathbf{k}) = \nabla_{\mathbf{k}} \times \boldsymbol{\mathcal{A}}_n(\mathbf{k}), \quad \boldsymbol{\mathcal{A}}_n(\mathbf{k}) = i \langle u_{n,\mathbf{k}}(\mathbf{x}) | \nabla_{\mathbf{k}} | u_{n,\mathbf{k}}(\mathbf{x}) \rangle \quad (2.1)$$

where $\boldsymbol{\mathcal{A}}_n$ is the Berry connection, and $|u_n\rangle$ is the periodic part of the Bloch wavefunction for band n . When a charge carrier is driven across k -space by an external laser field, the Berry curvature affects its intraband propagation within band n by introducing a lateral drift to its motion due to anomalous velocity, defined as:

$$\mathbf{v}_n(\mathbf{k}) = -\frac{e}{\hbar} \mathbf{F}(t) \times \boldsymbol{\Omega}_n(\mathbf{k}) \quad (2.2)$$

where $\mathbf{F}(t)$ is the driving field. The anomalous velocity mostly affects the evolution of the interaction in the polarization plane, due to phase matching considerations

[7, 8]. For a field propagating along z , while being polarized predominantly along y , the interaction is mostly affected by the x component of $\mathbf{v}_n$. Therefore, Ω_n^z , i.e. the z component of the Berry curvature, is considered as the main contribution affecting the intraband anomalous interaction.

The Berry curvature was calculated by DFT for each band and its z component is displayed in figure 4b along the $\Gamma - M$ and $\Gamma - K$ lines. As can be clearly observed, the Berry curvature can grow to large values along the $\Gamma - M$ symmetry axis. In contrast, along $\Gamma - K$ the Berry curvature is zero across all bands. This is a result of its anti-symmetric nature with respect to C_2 symmetry. The 2D Berry curvature is displayed in figure 4c for band 14, illustrating its angular dependence. Due to the C_2 symmetry, the Berry curvature flips its sign across the $\Gamma - K$ axis. In addition, due to the C_3 symmetry, the Berry curvature repeats itself after 120° rotation, with the Γ point acting as the center of rotation.

3 Interband mechanism in α - Quartz HHG

Since the first measurement, solid-state HHG [9] has become an active and rapidly growing field of research, providing a unique measurement scheme for studying ultra-fast dynamics in solids. However, while this field holds great promise, the underlying mechanism of the interaction is yet to be fully uncovered.

Several theoretical studies in recent years have addressed this challenge by solving the semiconductor Bloch equations (SBEs) for materials with large band-gap of several electron-Volts interacting with NIR fields. Comparing the contribution of interband and intraband HHG, these studies showed that the interband currents dominate the emission for above-band-gap harmonics [10, 11]. This mechanism is dominated by the interband transition dipole moment (TDM), which is defined for each two bands n, m and for every crystal momentum $\mathbf{k}$ as:

$$\mathbf{d}_{mn}(\mathbf{k}) = i \langle u_{m,\mathbf{k}}(\mathbf{x}) | \nabla_{\mathbf{k}} | u_{n,\mathbf{k}}(\mathbf{x}) \rangle \quad (3.1)$$

For a field polarized predominantly along the y direction, all interband transitions

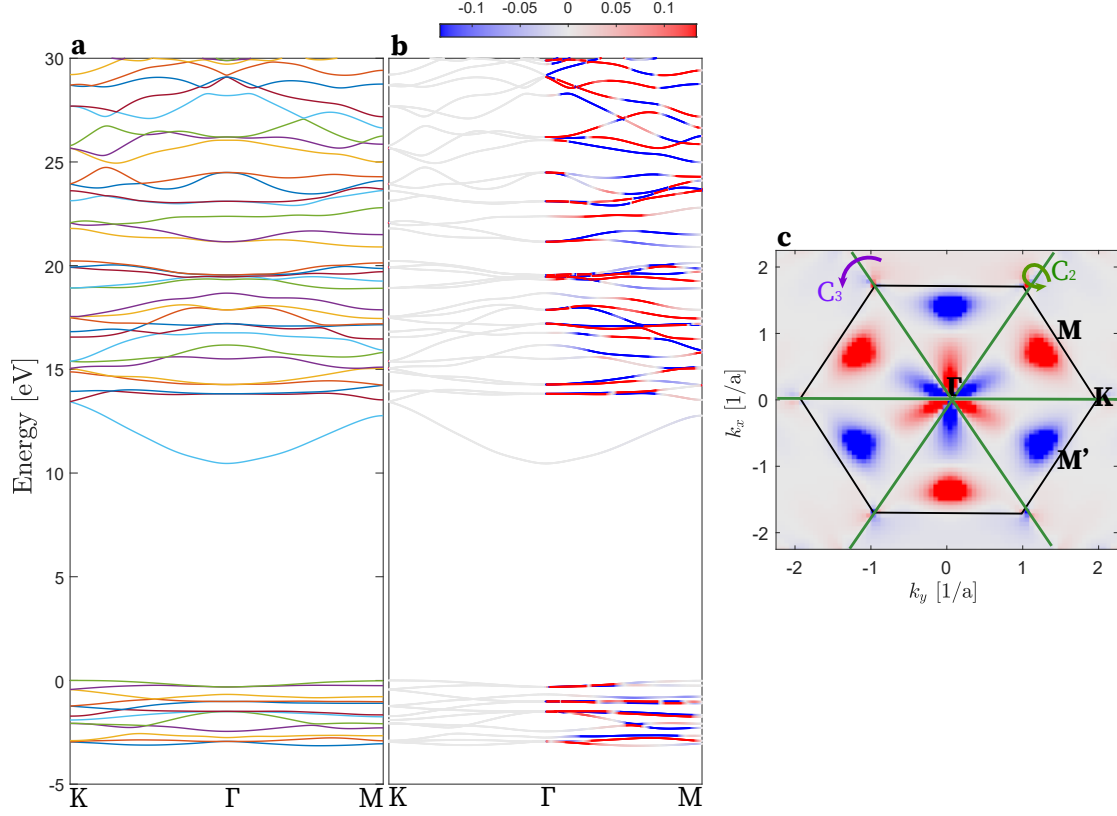


Figure 4: (a,b) α -quartz band structure along symmetry axes $K - \Gamma - M$ as extracted from DFT. In (a) each band is distinguished from neighboring bands by different colors. In (b) each band is color-coded with the band's Berry curvature's z component (parallel to field propagation direction) according to eq. 2.1 in main text. (c) The Berry curvature's z component of conduction band 14. The black dashed lines represent the edges of the first Brillouin zone. The golden lines represent C_2 axes.

occur primarily in parallel to the driving field's polarization, dictated by d_{mn}^y . This property plays an important role in various non-adiabatic transitions between the bands. Importantly it dictates the conduction-to-valence transition, which is at the heart of the recombination model.

Experiments in MgO, a strong insulator with 7.9eV minimal bandgap, have demonstrated HHG emission from several distinct conduction bands. These experiments revealed a strong link between bandgap and harmonic energy, showing unequivocally that the origin of emission is in electron-hole recombination, associated with the interband current [12]. In quartz, having a larger bandgap ($\sim 10\text{eV}$), the dominant role of the interband mechanism is expected to be exemplified even further. This is indeed supported by our data, as we will demonstrate hereafter.

Previous studies have shown a direct link between bright HHG spectral features to regions in the band structure which are characterized by strong interband dipole coupling or high density of states (DOS) [12]. In the following we perform a similar study in quartz. Figure 5(b,c) describes the band gap between each conduction band and the highest valence bands $\varepsilon_{c,n} - \varepsilon_{v,m}$, color-coded by the coupling strength. We mark the areas having strong coupling, as indication of larger recombination probabilities, as well as near-constant band gap, as signature of high DOS. Both conditions, put together, may indicate the enhancement of higher order harmonics, revealing the mapping between the band structure and harmonic spectrum, thus identifying the role of the interband mechanism.

Figure 5a depicts the HHG spectrum of Quartz up to 30eV , measured using a linearly polarized driving field along y direction with $\sim 5 \cdot 10^{13} \frac{\text{W}}{\text{cm}^2}$ intensity and 1200nm wavelength. The TDMs between conduction bands within that energy range and the two highest valence bands (11 and 12) were calculated by DFT (see chapter 2). The y -component of the TDM between each conduction band and band 11 (12) is displayed along their band gap in figure 5b(c). Due to the exponential dependence of the tunneling ionization mechanism with the band gap, we assume that the electron first tunnels to the lowest conduction band (band 13), followed by additional tunneling processes into higher conduction bands. Along $\Gamma - K$, the first conduction band is coupled to the lower valence band (band 11), hence it is the only band to be

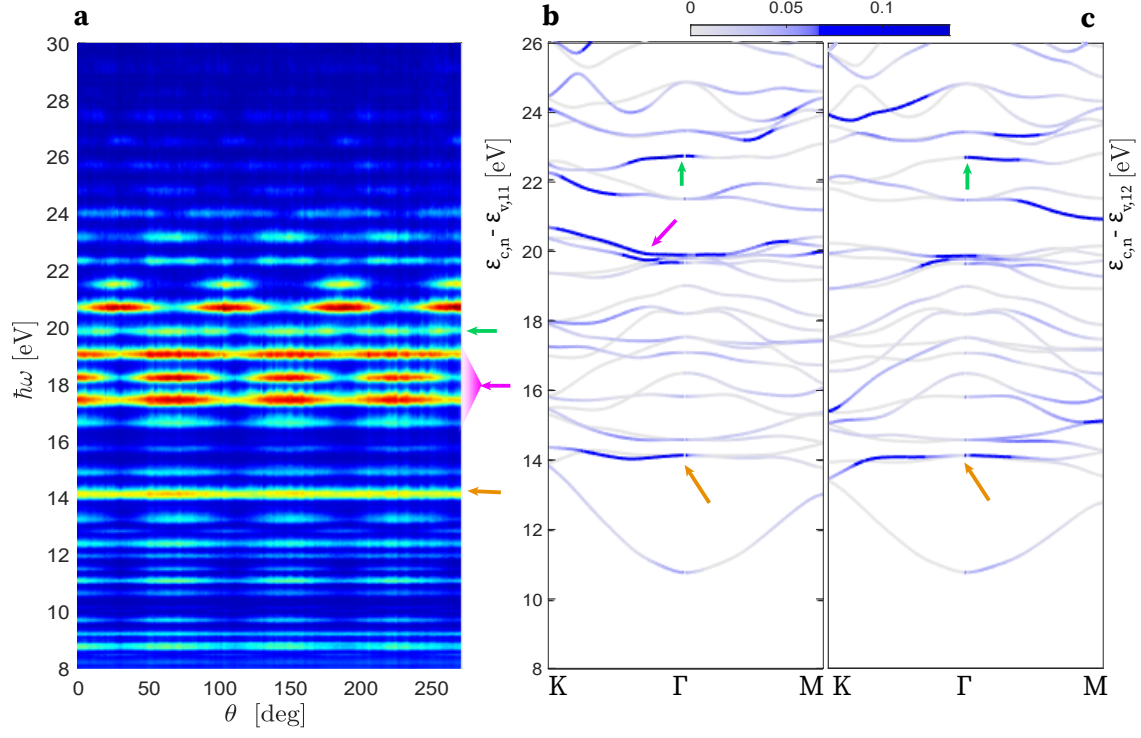


Figure 5: (a) Resolved HHG spectrum driven by linearly polarized field with central wavelength of 1200nm and $\sim 5 \cdot 10^{13} \frac{\text{W}}{\text{cm}^2}$ intensity. Harmonics 15, 20 and 22 are marked with orange, purple and green arrows, respectively. (b,c) Quartz band gap with respect to the two highest valence bands - bands 11, $\varepsilon_{c,n} - \varepsilon_{v,11}$ (b) and 12, $\varepsilon_{c,n} - \varepsilon_{v,12}$ (c), along symmetry axes $K - \Gamma - M$. The plots are color-coded with the TDM y-component amplitude (parallel to the field's main polarization axis) $d_{(c,n),11}$ (b) and $d_{(c,n),12}$ (c), respectively, according to eq. 3.1 in main text. The energy regions along the $\Gamma - K$ and $\Gamma - M$ lines corresponding to harmonics 15, 20 and 22 in (b) are marked by orange, purple and green arrows, respectively.

considered (figure 5b). In contrast, along $\Gamma - M$, both valence bands are coupled to the conduction band (band 12 having a stronger coupling), and are both strongly coupled to each other. Therefore we examine the coupling between conduction bands to both valence bands (figure 5b and c).

A comparison between the dipole couplings and the HHG spectrum (figure 5a) identifies several clear correlations. Around $19 - 21eV$ we observe a group of bands with a flat band region and strong coupling w.r.t band 11 along the $\Gamma - K$ axis (marked in purple). These bands can be directly linked to the enhanced emission of harmonics 19 – 21 at 0° , 60° and 120° , corresponding to the $\Gamma - K$ symmetry axis. In addition, at $15eV$ we notice a flat region band that is strongly coupled along both symmetry axes (marked in orange). Indeed, strong HHG emission is observed at harmonic 15 for both axes. Similarly, the emission of harmonic 22 at $\sim 22.5eV$ can be attributed to a strongly coupled band along both symmetry axes, resulting in enhanced emission over a wide angular range. It is noteworthy that bands 11 and 12 are strongly coupled to lower valence bands as well, so in principle the hole can tunnel between them much like the electron, and recombination can occur from high conduction bands to low valence bands, causing a spectral shift w.r.t to the conduction band's energy. However, this shift is expected to be rather small, since all valence bands are confined within a small energetic range of less than $3eV$ (figure 4a).

4 Semi classical trajectory analysis

In order to evaluate the role of Berry curvature in the electron-hole dynamics, we perform a semi-classical 2D trajectory simulation in real space. We calculate the electron-hole trajectories that evolve within a single cycle of the driving electric field, along the Γ -K or Γ -M directions. The driving field was set at a wavelength of $1.3[\mu m]$ and an intensity of $0.007[a.u.]$. Our calculations were performed using driving field with a wavelength of $1.2[\mu m]$ due to technical reasons only and we expect them to be very similar to $1.3[\mu m]$ driving field case. The band structure and Berry curvature are calculated via density functional theory (DFT) calculations (see fig. 7) with an

inner valence band chosen to avoid band crossings that affect the numerical error. The dynamics of the electron-hole pair is governed by the semi-classical equations of motion for solids (atomic units applied) [8]:

$$\langle \dot{\mathbf{k}}(t) \rangle = -\mathbf{F}(t) \quad (4.1)$$

$$\langle \mathbf{v}(t) \rangle = \nabla_{\mathbf{k}} \varepsilon_{cv}(\mathbf{k})|_{\mathbf{k}=\langle \mathbf{k}(t) \rangle} + \langle \dot{\mathbf{k}}(t) \rangle \times \boldsymbol{\Omega}_{cv}(\mathbf{k}(t)) \quad (4.2)$$

where ε_{cv} is the bandgap between conduction and valence bands, while $\boldsymbol{\Omega}_{cv}$ is the Berry curvature contribution of the electron (conduction) band and the hole (valence) band defined as:

$$\varepsilon_{cv}(\mathbf{k}) \equiv \varepsilon_c(\mathbf{k}) - \varepsilon_v(\mathbf{k}) \quad (4.3)$$

$$\boldsymbol{\Omega}_{cv}(\mathbf{k}) \equiv \boldsymbol{\Omega}_c(\mathbf{k}) - \boldsymbol{\Omega}_v(\mathbf{k}) \quad (4.4)$$

$\langle \mathbf{v}(t) \rangle$ in eq. (4.2) is the expectation value of the relative velocity operator between the electron and the hole. The first term on the right hand side of eq. (4.2) represents the relative group velocity calculated at specific momentum $\mathbf{k}$ in the band, while the second term is the anomalous velocity, governed by the Berry curvature.

Our analysis is based on the interband model [15]. Under the influence of the strong laser field an electron tunnels from the valence band into the conduction band, initiating an electron-hole wavepacket. Tunneling evolves at the Γ point in the Brillouin zone, where the crystal momentum is zero. As the electric field evolves, the electron-hole pair travels in the Brillouin zone, acquiring crystal momentum according to eq. (4.1). The crystal momentum is then translated into real-space displacement, dictated by the band's dispersion and the Berry curvature (eq. (4.2)). When the electron and hole wave-packets overlap in real space, recombination occurs, emitting a photon with energy equal to the energy difference between the conduction and the valence bands, at the instant of recombination:

$$\Delta r(t', t) = \int_{t'}^t v(\tau) d\tau = 0 \quad (4.5)$$

$$\varepsilon_{cv}(k(t)) = \hbar\omega \quad (4.6)$$

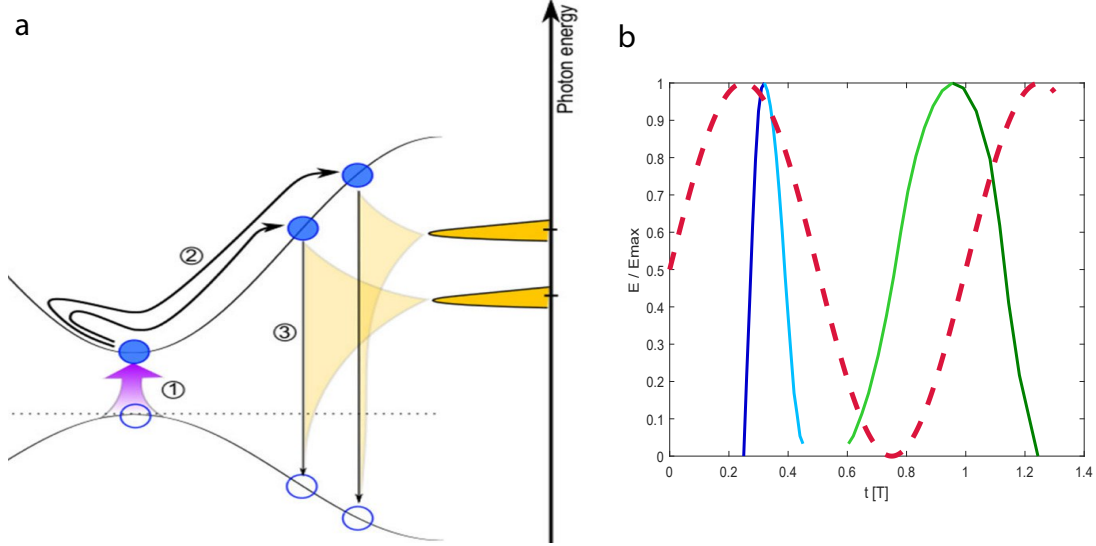


Figure 6: Time-Energy mapping: **(a)** Schematic description of interband mechanism: (1) an electron is tunnel ionized at time t' , leaving a hole in the valence band. Then, (2) both propagate, until (3) overlapping in real space and recombining at time t . The emitted photon energy is equal to the band gap energy at the recombination moment (picture taken from [13]). Different trajectories lead to the measured broad emission spectrum. **(b)** The ionization times (blue) and recombination times (green) of the different trajectories that lead to their corresponding emitted photon (y axis), in terms of the driving field period (dashed red). The light and dark colors distinguish between two families of trajectories, short and long [14]. In our analysis we consider the short trajectories only, characterised by later ionization times and shorter recombination times.

Where (t', t) are the tunneling and recombination times, $k(t)$ is the crystal momentum at the recombination moment and $\hbar\omega$ is the photon energy. Note that the recombination is induced when the electron and hole overlap in real space within the characteristic electron's wave-packet width. We evaluate the wave-packet's width by following the decay of the harmonics signal with the fundamental field's ellipticity. This width is defined by ellipticity at which the harmonics signal approaches FWHM level. We consider a set of possible tunneling times (t') , leading to different k space/real space trajectories and recombination events (t, ω) . This provides the mapping between the measured photon emission, resolved in the HHG spectrum, to the underlying k space dynamics (figure 6a)[16]. In figure 6b we present this calculated mapping, according to our semiclassical analysis described above, considering a linear field. This figure describes, for each photon energy, the ionization time and recombination time.

To fully evaluate the effect of the Berry curvature on the trajectories, we consider three different cases:

1. Single-color linearly-polarized light
2. Single-color elliptically-polarized light
3. Two-color cross-polarized light

In the following sections we provide a detailed analysis of these cases and extract their response to the Berry curvature.

4.1 Single-color linearly-polarized light

We perform the simulations for a single-color, linearly-polarized driving electric field ($A^y = A_0^y \sin(\omega_0 t)$), calculating the electron-hole dynamics in both half cycles. Figure 8 illustrates the real space trajectories evolving after the ionization step for two consecutive half cycles. Recombination occurs at the second time when the trajectory intersect with $y = 0$ (the relative distance between the electron and the hole vanishes). We focus on two important cases: i) the Γ -K direction, where the Berry curvature

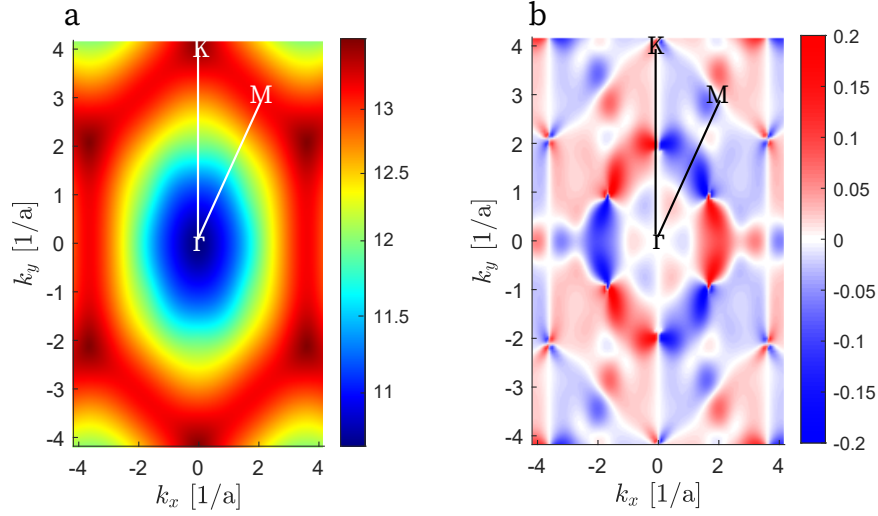


Figure 7: DFT calculation of **(a)** the energy gap ε_{cv} (units of eV) and **(b)** Berry curvature difference Ω_{cv} (units of $\frac{1}{a^2}$, a being the lattice constant) over the first Brillouin zone, for the lowest conduction band and an inner valance band.

vanishes; and ii) the Γ -M direction, where the Berry curvature is maximal. For the Γ -K direction, we observe an analogous behavior to an inversion-symmetric material: no transverse displacement (x direction, red line) occurs with respect to the field's polarization (y direction) (Fig. 8 top panel). In contrast, for the Γ -M direction, we observe a significant transverse displacement along the x direction. The directionality of this displacement is preserved between the two half cycles, since both the electric field and the Berry curvature invert their sign (Fig. 8 bottom panel).

4.2 Single-color elliptically-polarized light

In the next stage, we simulate the trajectories driven by a single-color elliptically-polarized light ($\epsilon A_0^y \cos(\omega_0 t), A_0^y \sin(\omega_0 t)$). In the absence of Berry curvature (i.e., along the Γ -K direction), the displacement of the electron-hole pair in the transverse direction (x) changes sign between two consecutive half cycles. In the presence of

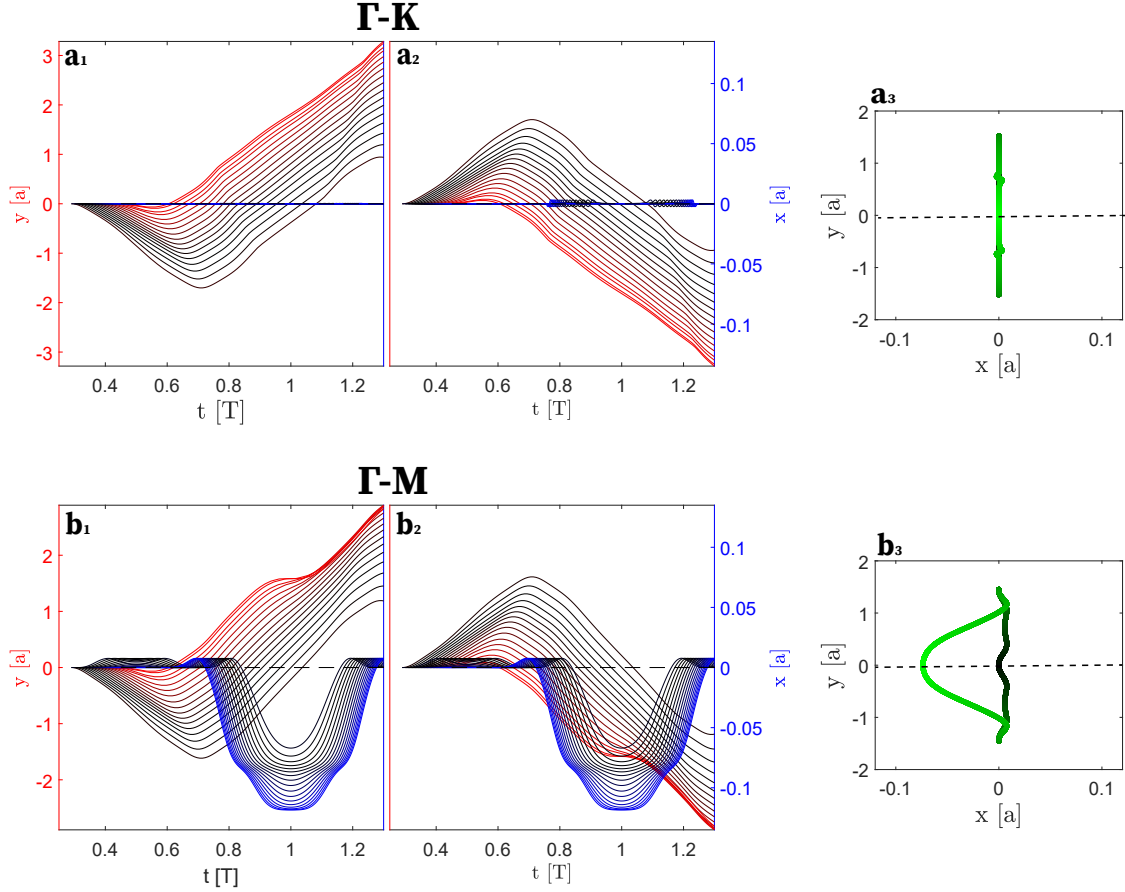


Figure 8: Electron-hole trajectories, driven by a single-color linearly-polarized light, along the Γ -K (upper panel, **a1:a2**) and Γ -M (lower panel, **b1:b2**) directions for each half-cycle of the field. **a1** (**a2**) and **b1** (**b2**) represent the trajectories along positive (negative) subcycle. The relative displacement between electron and hole is shown in units of lattice constant. The left axis (red) is the y displacement (along the fundamental field's polarization), while the right axis (blue) is the x displacement. The brightness of the different plots encodes the mapping to recombination energy: light (dark) colored trajectory will be map to low (high) recombination energy as well as to low (high) emitted photon energy. The trajectories are drawn beyond the recombination time to provide a schematic visualization of their response. **a3**, **b3** x-y cross-sectional view of a one chosen trajectory for Γ -K and Γ -M orientations, during the evolution along the two subcycles. The varying color encodes time from ionization (black) to recombination (green).

Berry curvature (i.e., along the Γ -M direction), for small epsilons, the trajectories of both half cycles stay similar, with only the accumulated phase being different due to the flip in the Berry curvature sign (see fig. 9).

4.3 Two-color cross-polarized light

Finally, we simulate the trajectories under the influence of a cross-polarized two-color light, $(\epsilon A_0^y \sin(2\omega_0 t + \varphi), A_0^y \sin(\omega_0 t),)$, where φ is the two fields subcycle delay. The second harmonic (SH) field amplitude is 5% of the fundamental field's amplitude. In this case, the transverse displacement is dictated by the presence of the SH field, which exerts a transverse force (i.e., in the x direction) as well as the Berry curvature. This force can either enhance or reduce the transversal displacement, induced by the anomalous velocity, *for both subcycles*. For each crystal orientation, Γ -K and Γ -M, we describe the trajectories, for two color delays of -0.5π and 0.5π (see fig. 10). For the Γ -K direction (no Berry curvature), the transverse displacement amplitude (x-direction, blue line) is invariant to an integer multiple of a π phase shift (see fig. 10, upper panel). Along this direction the Berry curvature vanishes, therefore no anomalous velocity arises. Scanning the two color delay will lead to a periodic modulation of the recombination signal, and therefore of the HHG signal, at twice the SH frequency. Conversely, for the Γ -M direction (maximal Berry curvature), the transverse displacement is suppressed for the -0.5π phase shift, but enhanced for the 0.5π phase shift (see fig. 10, lower panel). This observation occurs symmetrically for both half cycles, since the two-color field features the same symmetry between half-cycles, as it is for the anomalous velocity. Such response breaks the symmetry between $\tau = -0.5\pi$ and $\tau = 0.5\pi$, reducing the periodicity of the two color delay to be a full period of the SH field.

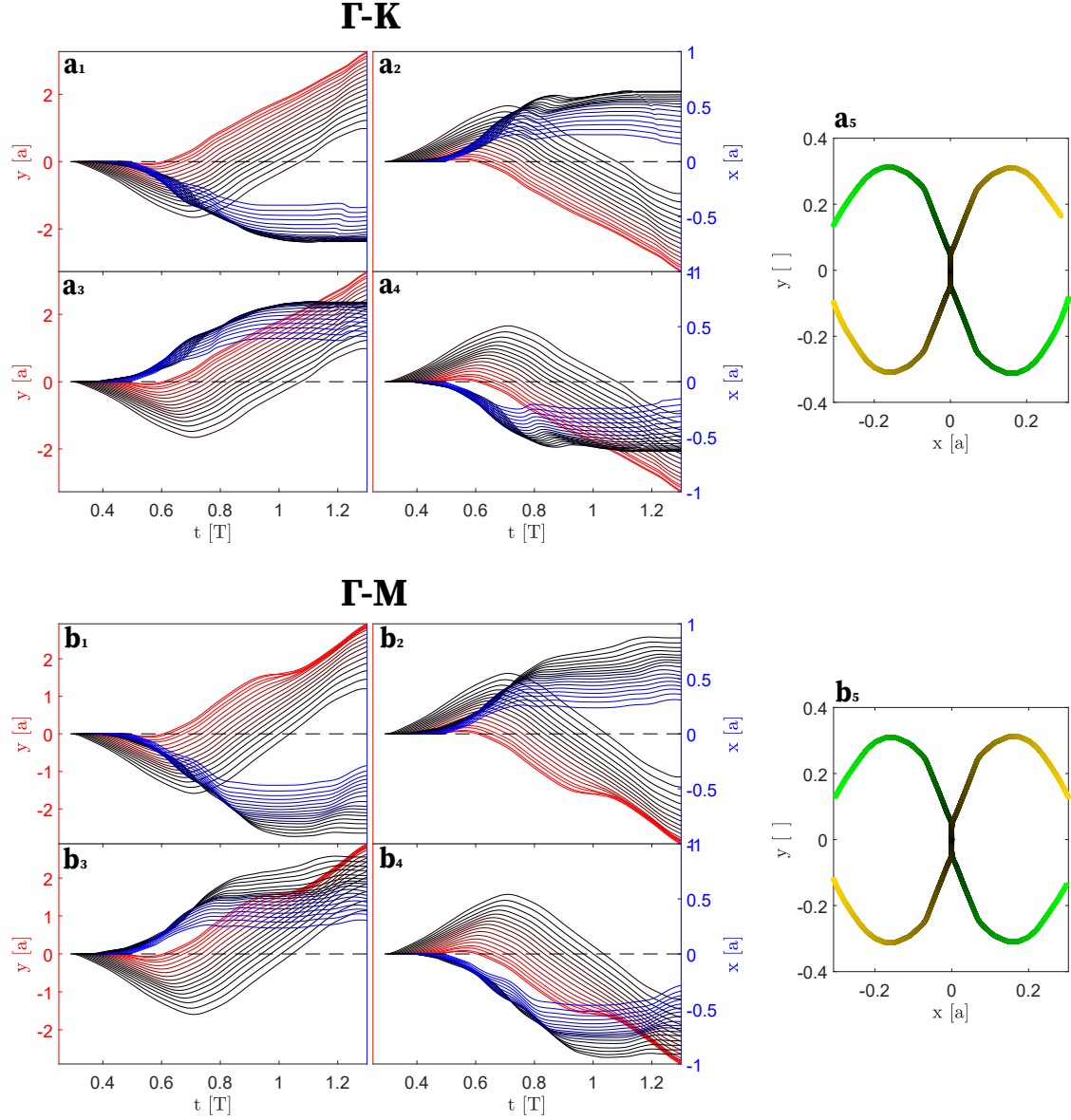


Figure 9: Electron-hole trajectories, driven by an elliptically polarized single-color field, along the Γ -K (upper plots) and Γ -M (lower plots) directions for $\epsilon = 0.1$ (**a1,a3, b1,b3**) and $\epsilon = -0.1$ (**a2,a4, b2,b4**) with each subplot representing a half-cycle of the field. **a1,b1** (**a2,b2**) and **a3,b3** (**a4,b4**) represent the trajectories during a positive (negative) subcycle. The relative displacement between electron and hole in units of lattice constant are shown: The left axis (red) is the y displacement while the right axis (blue) is the x displacement. The brightness of the different plots encodes the mapping to recombination energy. Light (dark) colored trajectories are mapped to low (high) recombination energies as well as to low (high) emitted photon energies. The trajectories are drawn beyond the recombination time to provide a schematic visualization of their response. **a5, b5** x-y plot of a single chosen trajectory for $\epsilon = \pm 0.1$ (green) and $\epsilon = -\pm 0.1$ (gold), during both subcycles. The brightness encodes the time evolution from ionization (dark) to recombination (light).

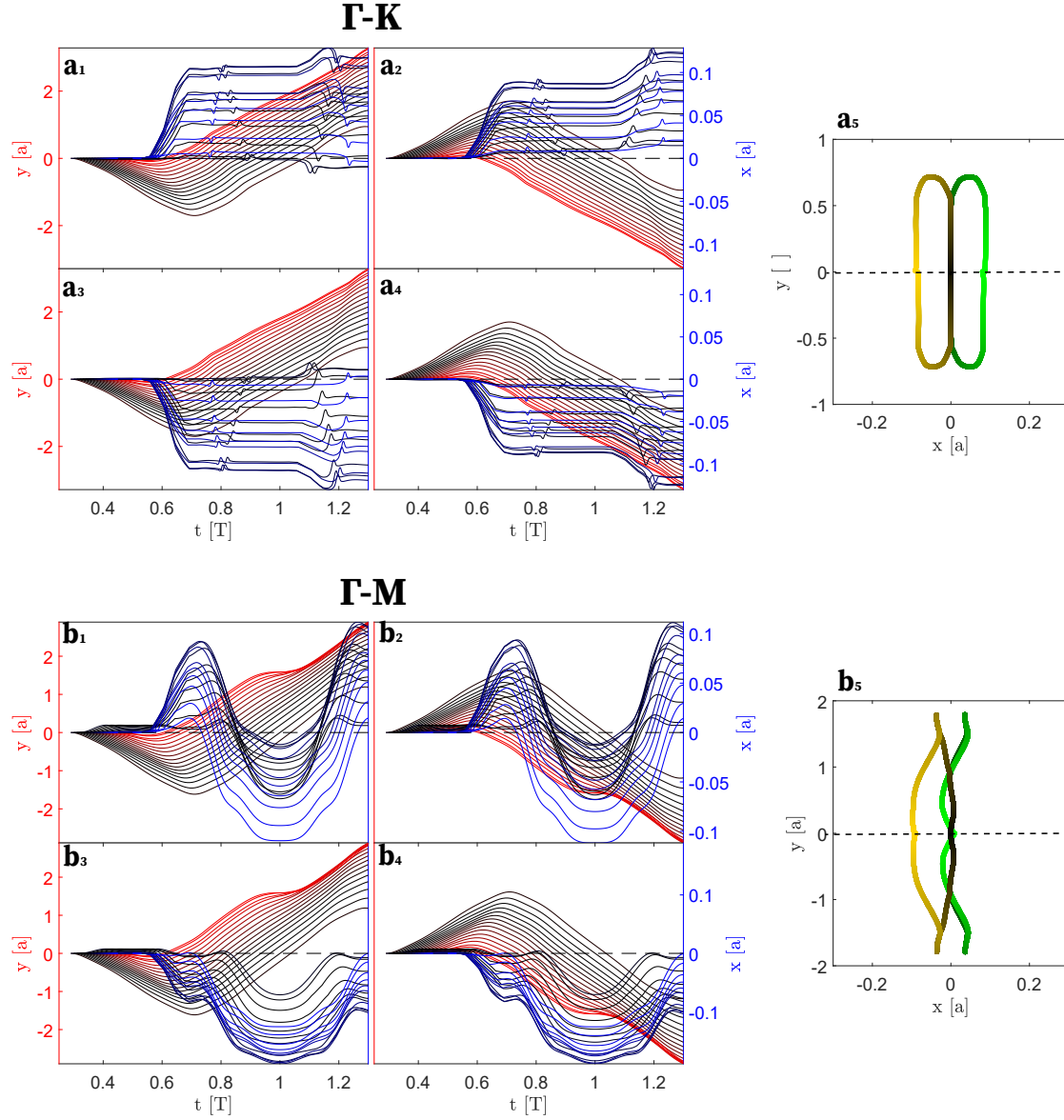


Figure 10: Electron-hole trajectories, driven by a two-color orthogonally-polarized field, along the Γ -K (upper panel) and Γ -M (lower panel) directions for $-\pi/2$ (**a1-a2**, **b1-b2**) and $\pi/2$ (**a3-a4**, **b3-b4**) two color delay. Each subplot represents a half-cycle of the field. **a1,b1** (**a2,b2**) and **a3,b3** (**a4,b4**) represent the trajectories during a positive (negative) subcycle. The relative displacement between electron and hole is shown in units of lattice constant. The left axis (red) is the y displacement, while the right axis (blue) is the x displacement. The brightness of the different plots encodes the mapping to recombination energy. Light (dark) colored trajectories are mapped to low (high) recombination energies as well as to low (high) emitted photon energies. The trajectories are drawn beyond the recombination time to provide a schematic visualization of their response. **a5**, **b5** x-y plot of a single chosen trajectory for $-\pi/2$ (green) and $\pi/2$ (gold) two color delays, during both subcycles. The brightness encodes the time evolution from ionization (dark) to recombination (light) time.

5 HHG Spectroscopy in broken inversion crystals- Quantum analysis

We start with a general description of HHG spectroscopy, resolved in a non-inversion symmetric crystal, adapted from ref [10]. We focus on the interband (recombination) picture, considering a two-band system. As was described in chapter 3, the interband mechanism is the dominant mechanism in our experimental conditions [10, 17]. While the following description considers a two-band system, we note that it can be easily extended to a multiband system[17]. Next, we present the semi-classical limit [14] of this quantum study, coinciding with the semi-classical analysis of Bloch electrons (chapter 4). Finally, we present a broad discussion concerning the interband Berry phase, as a manifestation of discrete geometrical phase. This includes its mathematical formalism and the relation to HHG spectroscopy.

5.1 Interband emission

A quantum description [14] of the inter-band mechanism is obtained by solving the Semiconductor Bloch Equations (SBE) using so-called Keldysh approach, introduced by Vampa et al. [15]. According to this approach, the interband current is dictated by a coherent sum over all quantum paths, weighted by their dipole coupling and the semi-classical action:

$$\mathbf{J}_{inter}(\omega) = i \int_{BZ} d^3K \int_{-\infty}^t dt' \int_{-\infty}^{\infty} dt \mathbf{F}(t') \cdot \mathbf{d}(\mathbf{K} + \mathbf{A}(t')) \mathbf{d}^*(\mathbf{K} + \mathbf{A}(t)) e^{-iS[\mathbf{K}, t', t] + i\omega t} + c.c \quad (5.1)$$

Where $S[\mathbf{k}, t', t]$ is the semi-classical action which in a broken inversion system contains the band dispersion as well as the Berry connection:

$$S[\mathbf{K}, t', t] \equiv \int_{t'}^t [\varepsilon_g(\mathbf{K} + \mathbf{A}(\tau)) + \mathbf{F}(\tau) \cdot \mathcal{A}_g(\mathbf{K} + \mathbf{A}(\tau))] d\tau \quad (5.2)$$

Here $\mathbf{d}(\mathbf{k}) = i \langle u_{v,\mathbf{k}}(x) | \nabla_{\mathbf{k}} | u_{c,\mathbf{k}}(x) \rangle$ is the dipole coupling between the valence and

the conduction bands, $u_{v/c,\mathbf{k}}(x)$ is the periodic part of the Bloch functions ($\psi_{n,\mathbf{k}} = e^{i\mathbf{k}\cdot\mathbf{r}}u_{n,\mathbf{k}}(r)$) and $\varepsilon_g = \varepsilon_c - \varepsilon_v$ is the two bands energy difference. $\mathcal{A}_g = \mathcal{A}_c - \mathcal{A}_v$ is their relative Berry connection, defined as $\mathcal{A}_m(\mathbf{k}) = i \langle u_{m,\mathbf{k}}(x) | \nabla_{\mathbf{k}} | u_{m,\mathbf{k}}(x) \rangle$, in which $m = v, c$. $\mathbf{F}(t)$ and $\mathbf{A}(t)$ are the laser field and its vector potential and $\mathbf{K}(\mathbf{t}) = \mathbf{k} - \mathbf{A}(t)$ is the canonical crystal quasi-momentum.

The system symmetries dictate the complex properties of the dipole coupling. Time reversal symmetry leads to $\mathbf{d}(-\mathbf{k}) = \mathbf{d}^*(\mathbf{k})$ and inversion symmetry to $\mathbf{d}(-\mathbf{k}) = -\mathbf{d}(\mathbf{k})$. Therefore, the dipole coupling induced in a crystal with inversion symmetry is purely imaginary. However, in a broken inversion crystal, the dipole coupling is a complex quantity, composed by real and imaginary components. Adding the dipole phases to the action, defining the *generalized action*, provides a gauge invariant quantity, as presented in the main text (Equation 1):

$$J_{inter}^{(i)}(\omega) = i \int_{BZ} d^3K \int_{-\infty}^t dt' \int_{-\infty}^{\infty} dt F^{(j)}(t') \cdot |d^{(j)}(\mathbf{K} + \mathbf{A}(t'))| |d^{*(i)}(\mathbf{K} + \mathbf{A}(t))| e^{-iS^{(i)}[\mathbf{K}, t', t] + i\omega t} + c.c$$

$$S^{(i)}[\mathbf{K}, t', t] \equiv \int_{t'}^t [\varepsilon_g(\mathbf{K} + \mathbf{A}(\tau)) + \mathbf{F}(\tau) \cdot \mathcal{A}_g(\mathbf{K} + \mathbf{A}(\tau))] d\tau + \phi_d^{(j)}(\mathbf{K} + \mathbf{A}(t')) - \phi_d^{(i)}(\mathbf{K} + \mathbf{A}(t)) \quad (5.3)$$

Where ϕ_d is the dipole coupling phase and $j, i = x, y$. We separate the action to its trivial component, which controls the interaction in inversion symmetric crystal and to the gauge invariant Berry phase:

$$S^{(i)}[\mathbf{K}, t', t] \equiv S_{inv} + \gamma_B^{(i)}$$

$$S_{inv}[\mathbf{K}, t', t] \equiv \int_{t'}^t \varepsilon_g(\mathbf{K} + \mathbf{A}(\tau)) d\tau \quad (5.4)$$

$$\gamma_B^{(i)}[\mathbf{K}, t', t] \equiv \int_{t'}^t \mathbf{F}(\tau) \cdot \mathcal{A}_g(\mathbf{K} + \mathbf{A}(\tau)) d\tau + \phi_d^{(j)}(\mathbf{K} + \mathbf{A}(t')) - \phi_d^{(i)}(\mathbf{K} + \mathbf{A}(t))$$

Duo to time reversal symmetry, S_{inv} is symmetric at positive and negative crystal momentum, $S_{inv}(-\mathbf{k}) = S_{inv}(\mathbf{k})$, while the Berry phase switches its sign, $\gamma_B(-\mathbf{k}) = -\gamma_B(\mathbf{k})$. Finally, in order to simplify the time dependence of this expression we

rewrite the Berry phase as:

$$\begin{aligned}
\gamma_B^{(i)}[\mathbf{K}, t', t] &\equiv \tilde{\gamma}_B + \Delta\phi_d^{(i)} \\
\tilde{\gamma}_B &\equiv \int_{t'}^t \mathbf{F}(\tau) \cdot [\mathcal{A}_g(\mathbf{K} + \mathbf{A}(\tau)) + \nabla_K \phi_d^{(j)}(\mathbf{K} + \mathbf{A}(\tau))] d\tau \\
\Delta\phi_d^{(i)} &\equiv \phi_d^{(j)}(\mathbf{K} + \mathbf{A}(t)) - \phi_d^{(i)}(\mathbf{K} + \mathbf{A}(t))
\end{aligned} \tag{5.5}$$

Note that the Berry phase contains the difference between the recombination dipole phase of the two polarization components, which becomes zero when the dipole is parallel to the fundamental field's polarization. Therefore, it is useful to define the polarization independent Berry phase, $\tilde{\gamma}_B$, which is mutual at both polarization components.

5.2 Saddle point approximation

Equation (5.3) provides an expression for the interband current generated in the HHG process, resembling the three steps in the three-step model: (i) first, the electron is tunnel-ionized by the electric field $F^{(j)} d^{(j)}(\mathbf{K} + \mathbf{A}(t'))$ accumulating the ionization dipole phase, (ii) then, the electron propagates inside the bands, accumulating a phase $e^{-iS[\mathbf{K}, t', t]}$ (iii) finally, the electron recombines with its parent hole $d^{*(i)}(\mathbf{K} + \mathbf{A}(t))$, accumulating the recombination dipole phase. The recombination leads to the emission of a photon $e^{-i\omega t}$, having an energy dictated by the associated electron-hole energy gap. All quantum paths are then coherently summed over, each one corresponding to different ionization times and ionization locations in the Brillouin zone $\int_{BZ} d^3\mathbf{K} \int_{-\infty}^t dt'$. Their interference dictates the final interband current as well as the spectrum of the emitted HHG.

An intuitive physical picture of the underlying dynamics can be achieved by taking an approximation, known as the Saddle Point Approximation (SPA). When $e^{-iS[\mathbf{K}, t', t]}$ oscillates rapidly compared to the pre-exponential factors, then only quantum paths near the stationary points of the action S contribute to the interband current. These paths are defined by a set of parameters $\{\mathbf{K}_{st}, t'_{st}, t_{st}\}$ for which $\nabla S = 0$, which are

derived from the following stationary equations [10, 18]:

$$\varepsilon_g(\mathbf{K} + \mathbf{A}(t'_{st})) + \mathbf{F}^{(j)}(t'_{st}) \mathcal{D}^{(j)}(t'_{st}) = 0 \quad (5.6)$$

$$\Delta \mathbf{r} - \mathcal{D}^{(i)}(t_{st}) + \mathcal{D}^{(j)}(t'_{st}) = \mathbf{0} \quad (5.7)$$

$$\varepsilon_g(\mathbf{K} + \mathbf{A}(t_{st})) + \mathbf{F}(t_{st}) \cdot [\mathcal{D}^{(j)}(t'_{st}) + \Delta \mathbf{r}] = \omega \quad (5.8)$$

with (j) and (i) being the components parallel and perpendicular to the ionization field respectively, and:

$$\Delta \mathbf{r} \equiv \int_{t'_{st}}^{t_{st}} [\mathbf{v}_c(\tau) - \mathbf{v}_v(\tau)] d\tau \quad (5.9)$$

$$\mathbf{v}_n(\tau) \equiv \nabla_{\mathbf{K}} \varepsilon_n(\mathbf{K} + \mathbf{A}(\tau)) - \mathbf{F}(\tau) \times \boldsymbol{\Omega}_n(\mathbf{K} + \mathbf{A}(\tau)) \quad (5.10)$$

$$\mathcal{D}^{(i)}(\tau) \equiv \mathcal{A}_g(\mathbf{K} + \mathbf{A}(\tau)) - \nabla_{\mathbf{K}} \phi_d^{(i)}(\mathbf{K} + \mathbf{A}(\tau)) \quad (5.11)$$

Eq. (5.10) defines two contributions to the wavepacket velocity: the band dispersion $\nabla_{\mathbf{K}} \varepsilon_n$, and the anomalous velocity $\mathbf{F} \times \boldsymbol{\Omega}_n$ due to Berry curvature, being perpendicular to the driving field.

In the following step we neglect the dipole phase contributions $\mathcal{D}^{(j)}(\tau)$, which are commonly small [18], simplifying the stationary equations to be:

$$\varepsilon_g(\mathbf{K} + \mathbf{A}(t'_{st})) = 0 \quad (5.12)$$

$$\Delta \mathbf{r} = 0 \quad (5.13)$$

$$\varepsilon_g(\mathbf{K} + \mathbf{A}(t_{st})) = \omega \quad (5.14)$$

According the first equation the electrons are born with zero crystal momentum, at time t'_{st} for zero band gap energy. The finite band gap energy results in a complex birth time, which represents the tunnel ionization process. The second equation reflects that recombination occurs when the electron and hole wavepackets overlap in real-space. Finally, the third stationary equation represents conservation of energy: the electron-hole pair recombines and emits a photon ω with an energy equal to the

energy band gap at moment of recombination t_{st} .

These results are fully compatible with the semi-classical equations of motion (4.2). The simulation recombination condition and emitted photon energy comply with equations (5.13) and (5.14), respectively. The quantum nature of HHG is captured by equation (5.12), where the tunneling process leads to an imaginary ionization time. The coupling between the different saddle point equations leads to a complex crystal momentum and recombination time as well as to a complex Berry phase.

5.3 Discrete Berry phase

In the following chapter we present a general description of discrete geometrical phase and its physical manifestation as interband Berry phase. A simple way to obtain a Berry phase is to take N states $|\Psi_i\rangle$ and to consider the phase of

$$\frac{\langle\Psi_1|\Psi_2\rangle\langle\Psi_2|\Psi_3\rangle\cdots\langle\Psi_N|\Psi_1\rangle}{|\langle\Psi_1|\Psi_2\rangle\langle\Psi_2|\Psi_3\rangle\cdots\langle\Psi_N|\Psi_1\rangle|} = e^{i\gamma} \quad (5.15)$$

(for a review, see for example [19], section 2.1), as shown in Fig. 11. The phase γ is gauge invariant in the sense that it is independent of the unphysical phase of each of the states $|\Psi_i\rangle$. This discrete description is quite general, and the familiar uses of the Berry phase are simply limits of this case.

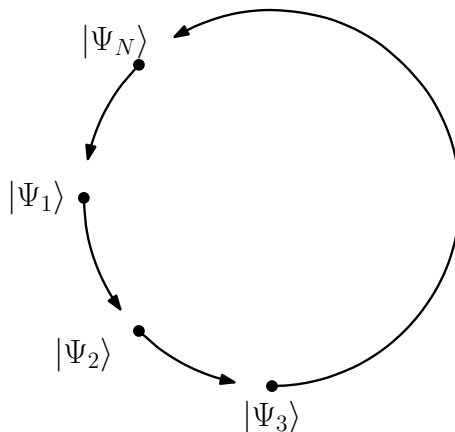


Figure 11: The accumulated Berry phase in a closed trajectory.

For example, in band theory the wavefunctions are Bloch waves $\psi_{n,k} = e^{ik \cdot r} u_{n,k}(r)$, and taking a trajectory in k space, (5.15) becomes

$$\langle u_{n,k_1} | u_{n,k_2} \rangle \langle u_{n,k_2} | u_{n,k_3} \rangle \cdots \langle u_{n,k_N} | u_{n,k_1} \rangle. \quad (5.16)$$

For nearby momenta, $\langle u_{n,k} | u_{n,k+\epsilon} \rangle \approx 1 + \epsilon \langle u_{n,k} | \partial_k u_{n,k} \rangle \approx e^{\epsilon \langle u_{n,k} | \partial_k u_{n,k} \rangle}$ with $\langle u_{n,k} | \partial_k u_{n,k} \rangle$ being purely imaginary. For infinitesimal ϵ , we find

$$\langle u_{n,k_1} | u_{n,k_2} \rangle \langle u_{n,k_2} | u_{n,k_3} \rangle \cdots \langle u_{n,k_N} | u_{n,k_1} \rangle \approx e^{\epsilon \sum_i \langle u_{n,k_i} | \partial_k u_{n,k_i} \rangle} \rightarrow e^{\int dk \langle u_{n,k} | \partial_k u_{n,k} \rangle} \equiv e^{-i \int dk \mathcal{A}_n(k)} \quad (5.17)$$

which is the familiar continuum Berry phase in a band, and $\mathcal{A}_n(k)$ is the Berry connection.

Interestingly, in High Harmonic Generation, we obtain a trajectory that goes between different bands. This trajectory is closed, see Fig. 12a. The finite (discrete) version of Fig. 12a is shown in Fig. 12b, and the corresponding expression (5.15) is just

$$\begin{aligned} & \langle u_v(k_1) | u_c(k_i) \rangle [\langle u_c(k_i) | u_c(k_1) \rangle \cdots \langle u_c(k_{N-1}) | u_c(k_N) \rangle] \cdot \\ & \cdot \langle u_c(k_N) | u_v(k_f) \rangle [\langle u_v(k_f) | u_v(k_N) \rangle \cdots \langle u_v(k_2) | u_v(k_1) \rangle] \\ & \approx \epsilon^2 \langle \partial_k u_v(k_i) | u_c(k_i) \rangle e^{\int_{k_i}^{k_f} dk \langle u_c(k) | \partial_k u_c(k) \rangle} \langle u_c(k_f) | \partial_k u_v(k_f) \rangle e^{-\int_{k_i}^{k_f} dk \langle u_v(k) | \partial_k u_v(k) \rangle} \end{aligned} \quad (5.18)$$

where we note that $\langle u_v(k) | u_c(k) \rangle = 0$ so that for $k_1 = k_i + \epsilon$ close to each other, we have $\langle u_v(k_1) | u_c(k_i) \rangle \approx (\langle u_v(k_i) | + \epsilon \langle \partial_k u_v(k_i) |) | u_c(k_i) \rangle = \epsilon \langle \partial_k u_v(k_i) | u_c(k_i) \rangle$. The continuous part of the trajectory goes from k_i to k_f . To summarize, the description of the wavefunction evolution using the Berry phase formalism (5.15) indeed gives us the familiar High Harmonic Generation expression

$$\begin{aligned} & \propto d^*(k_i) e^{-i \int_{k_i}^{k_f} dk \mathcal{A}_c(k)} d(k_f) e^{i \int_{k_i}^{k_f} dk \mathcal{A}_v(k)} = |d^*(k_i)| |d(k_f)| e^{-i \left(\int_{k_i}^{k_f} dk \mathcal{A}_c(k) - \mathcal{A}_v(k) + \phi_d(k_f) - \phi_d(k_i) \right)} \\ & \gamma \equiv \int_{k_i}^{k_f} dk [\mathcal{A}_c(k) - \mathcal{A}_v(k)] + \phi_d(k_f) - \phi_d(k_i) \end{aligned} \quad (5.19)$$

The discrete jump is described by the dipole moment, $d(k) = |d(k)|e^{-i\phi_d}$, which is a connection between the bands, similarly to the more standard connection, $\mathcal{A}_n(k)$ that goes within a band.

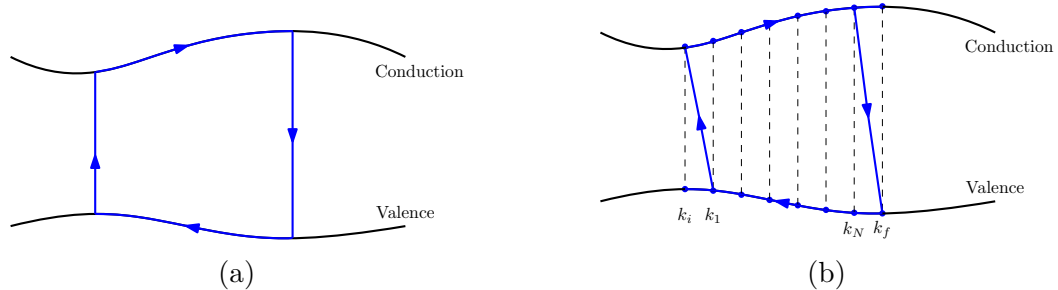


Figure 12: A closed trajectory passing through several bands.

The discrete part of the trajectory does not allow us to write the Berry phase as a surface integral over a gauge invariant curvature in a straightforward way. This is not unusual, as there are many cases where a Berry phase cannot be written in such a way. However, we can find a quantity, that we denote by $\mathbf{F}(k)$, similar to the Berry curvature ($\Omega(k)$), representing the flux over an infinitesimal area, and is gauge invariant. It is given by

$$\mathbf{F}(k) = \mathcal{A}_c(k) - \mathcal{A}_v(k) + \nabla_k \phi_d(k). \quad (5.20)$$

It can be verified that $F(k)$ is invariant under the gauge transformation

$$\begin{aligned} |u_c(k)\rangle &\rightarrow e^{i\alpha(k)} |u_c(k)\rangle, \\ |u_v(k)\rangle &\rightarrow e^{i\beta(k)} |u_v(k)\rangle. \end{aligned} \quad (5.21)$$

Then, we can write the Berry phase (5.18) as a line integral over $F(k)$

$$\gamma = \int_{k_i}^{k_f} d\mathbf{k} \cdot \mathbf{F}(\mathbf{k}) \quad (5.22)$$

Here $\mathbf{F}(k)$ represents a narrow rectangle in energy-momentum space and line integration over all these rectangles gives the total surface captured by the Berry

phase, see Fig. 13.

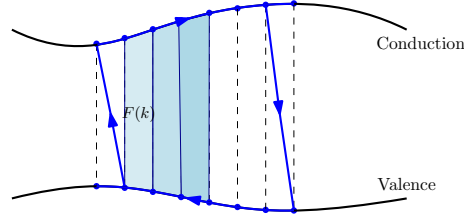


Figure 13: Integrated flux.

6 α -quartz HHG spectroscopy

6.1 Linearly Polarized Field

In the first stage we consider a single, multicycle field, having a linear polarization ($A^y(t) = A_0^y \sin(\omega_0 t)$). In this case, the interband mechanism leads to the emission of optical radiation every half of an optical cycle of the driving field, forming a train of identical attosecond pulses. The emission during the positive and negative half cycles is associated with electronic trajectories driven along the positive and negative crystal momentum, respectively. Summing these two contributions coherently, can be expressed as:

$$\begin{aligned} \mathbf{E}_{HHG}(\omega) = & \sum_{N=-\infty}^{\infty} i \int_{BZ} d^3K \int_{-\infty}^t dt' \int_0^{\frac{T}{2}} dt (\mathbf{F}(t') \cdot \mathbf{d}(\mathbf{K} + \mathbf{A}(t')) \mathbf{d}^*(\mathbf{K} + \mathbf{A}(t)) e^{-iS[\mathbf{K}, t', t] + i\omega t} \\ & - \mathbf{F}(t') \cdot \mathbf{d}(-\mathbf{K} - \mathbf{A}(t')) \mathbf{d}^*(-\mathbf{K} - \mathbf{A}(t)) e^{-iS[-\mathbf{K}, t', t] + i\omega t + iN\omega_0 \frac{T}{2}} + c.c) \delta(\omega - N\omega_0) \end{aligned} \quad (6.1)$$

Where $\mathbf{E}_{HHG}$ is the harmonic field, N is the harmonic number, T and ω_0 are the fundamental field's period and frequency, respectively. According to the time reversal symmetry (TRS), $\mathbf{d}(-\mathbf{k}) = \mathbf{d}^*(\mathbf{k})$, and based on the symmetry of the generalized

action, odd and even harmonic signals can be described as:

$$\begin{aligned}
E^{(i)}(N_{odd}) &\propto i \int_{BZ} d^3K \int_{-\infty}^t dt' \int_0^{\frac{T}{2}} dt F^{(j)}(t') |d^{(j)}(\mathbf{K} + \mathbf{A}(t'))| |d^{*(i)}(\mathbf{K} + \mathbf{A}(t))| e^{-iS_{inv}[\mathbf{K}, t', t] + i\omega t} \\
&\quad \cdot \cos(\gamma_B^{(i)}[\mathbf{K}, t', t]) + c.c \\
E^{(i)}(N_{even}) &\propto i \int_{BZ} d^3K \int_{-\infty}^t dt' \int_0^{\frac{T}{2}} dt F^{(j)}(t') |d^{(j)}(\mathbf{K} + \mathbf{A}(t'))| |d^{*(i)}(\mathbf{K} + \mathbf{A}(t))| e^{-iS_{inv}[\mathbf{K}, t', t] + i\omega t} \\
&\quad \cdot \sin(\gamma_B^{(i)}[\mathbf{K}, t', t]) + c.c
\end{aligned} \tag{6.2}$$

Equation 6.2 captures the interferometric nature of our measurements: the odd and the even harmonics encode constructive and destructive interference of the Berry phase, as their intensity presented in equation 2 at the main text.

By using Saddle Point Approximation, we estimate the integral above according to the stationary parameters of the action ($[\mathbf{K}^{st}, t'^{st}, t^{st}]$) [16]. Driving the interaction by a linearly polarized field (along y) the intensities of odd and even harmonics can be expressed as:

$$I^{(N)} \propto \begin{cases} g_{ion} e^{-2Im(S_{inv})} \left(|d^y|^2 |\cos \tilde{\gamma}_B|^2 + |d^x|^2 |\cos(\tilde{\gamma}_B + \Delta\phi_d^{(x)})|^2 \right) & N \text{ is odd} \\ g_{ion} e^{-2Im(S_{inv})} \left(|d^y|^2 |\sin \tilde{\gamma}_B|^2 + |d^x|^2 |\sin(\tilde{\gamma}_B + \Delta\phi_d^{(x)})|^2 \right) & N \text{ is even} \end{cases} \tag{6.3}$$

Where g_{ion} is the ionization probability, determined by the field's amplitude and the dipole coupling at the stationary tunneling time and momentum. d^x, d^y are the recombination dipole components, estimated by the stationary recombination time and momentum. Finally, S_{inv} and $\tilde{\gamma}_B$ are determined at the stationary parameters as well (note that these are complex quantities)

In the following we focus on the $\Gamma - M$ and $\Gamma - K$ symmetry axes. Quartz has a C_2 symmetry along the $\Gamma - K$ direction. Therefore, when the laser is polarized along the $\Gamma - M$ direction (i.e in perpendicular to the C_2 symmetry axis), the dipoles will

transform as:

$$\begin{cases} d_{mn}^x(-k_x, k_y) = s_m s_n d_{mn}^x(k_x, k_y) \\ d_{mn}^y(-k_x, k_y) = -s_m s_n d_{mn}^y(k_x, k_y) \end{cases} \quad (6.4)$$

where $s_m, s_n \in \{\pm 1\}$ are the parity eigenvalues of the Bloch wavefunction [20, 21]. For the first conduction band and the highest coupled valence band, denoted c and v , it holds that $s_c = s_v = 1$. In this case, d_{cv}^y is purely imaginary ($\phi_d^{(y)} = \frac{\pi}{2}$) and d_{cv}^x is purely real ($\phi_d^{(x)} = 0$), for all crystal momentum values, concluding that $\Delta\phi_d^{(x)} = \frac{\pi}{2}$. Such symmetries results in:

$$I^{(N)} \propto \begin{cases} e^{-2Im(S_{inv})} (|d^y|^2 |\cos \tilde{\gamma}_B|^2 + |d^x|^2 |\sin \tilde{\gamma}_B|^2) & N \text{ is odd} \\ e^{-2Im(S_{inv})} (|d^y|^2 |\sin \tilde{\gamma}_B|^2 + |d^x|^2 |\cos \tilde{\gamma}_B|^2) & N \text{ is even} \end{cases} \quad (6.5)$$

Additionally, along the $\Gamma - M$ direction, it holds that $\mathcal{A}^y(-k_y, 0) = -\mathcal{A}^y(k_y, 0)$ due to the C_2 symmetry in the perpendicular direction. In addition, $\mathcal{A}_g(-\mathbf{k}) = \mathcal{A}_g(\mathbf{k})$ due to TRS. Combining the two symmetries, we conclude that along this direction $\mathcal{A}^y = 0$ and therefore $\tilde{\gamma}_B = 0$. Eq. 6.5 is therefore reduced to:

$$I^{(N)} \propto \begin{cases} e^{-2Im(S_{inv})} |d^y|^2 & N \text{ is odd} \\ e^{-2Im(S_{inv})} |d^x|^2 & N \text{ is even} \end{cases} \quad (6.6)$$

In contrast, when the field is linearly polarized along the $\Gamma - K$ direction, it holds that $d^x = 0$ [7], therefore relation 6.3 becomes:

$$I^{(N)} \propto \begin{cases} e^{-2Im(S_{inv})} (|d^y|^2 |\cos \tilde{\gamma}_B|^2) & N \text{ is odd} \\ e^{-2Im(S_{inv})} (|d^y|^2 |\sin \tilde{\gamma}_B|^2) & N \text{ is even} \end{cases} \quad (6.7)$$

6.2 Elliptically Polarized Field

In this chapter we study HHG spectroscopy using elliptically polarized light, $\mathbf{A}(t) = A_0(\epsilon \cos(\omega_0 t), \sin(\omega_0 t))$, while controlling the ellipticity degree of the light, ϵ . As in the case of linear polarization, the instantaneous field reverses the order between two consecutive half cycles, therefore the temporal symmetry is preserved and eq.

(6.3) remains valid. We focus on the perturbative limit, $\epsilon \ll 1$, where the main parameters of the interaction are unchanged, while the ellipticity of the field induces a small lateral drift. Specifically, we use the unperturbed stationary parameters along the main polarization axis, $(K_y^{st}, t'^{st}, t^{st})$, adding the transversal evolution by ϵK_x^{st} , $(\epsilon K_x^{st}, K_y^{st}, t'^{st}, t^{st})$. We expand the action with ϵ as:

$$S^{(i)}[\mathbf{K}, t', t] \approx S_0^{(i)} + \frac{\partial S^{(i)}}{\partial \epsilon} \epsilon = S_{inv,0} + \gamma_{B,0}^{(i)} + \frac{\partial S_{inv}}{\partial \epsilon} \epsilon + \frac{\partial \gamma_B^{(i)}}{\partial \epsilon} \epsilon + \dots \quad (6.8)$$

Where $S_{inv,0}$ and $\gamma_{B,0}^{(i)}$ represent the unperturbed action for linear polarized field. We focus on the main axes, $\Gamma - K$ and $\Gamma - M$. First, we analyse the symmetric part of the action, S_{inv} . Its dependence on ϵ can be expressed as: $\frac{\partial S_{inv}}{\partial \epsilon} = \frac{\partial S_{inv}}{\partial K_x} K_x^{st}$. Along the main axes, the first order term in the expansion vanishes, therefore only second order terms contribute. We define the ellipticity response of this term as $\Delta S_{inv} \equiv \Delta \varepsilon_g \equiv \frac{\partial S_{inv}}{\partial^2 K_x^{st}} \epsilon^2 K_x^{st}$ (see in the main text). However, the Berry phase leads to a first order contribution, dictated by the transverse crystal momentum as well as the transverse field:

$$\begin{aligned} \frac{\partial \gamma_B^{(i)}}{\partial \epsilon} &\approx \frac{\partial \tilde{\gamma}_B}{\partial \epsilon} \propto \int_{t'}^t \left[\frac{\partial(F^{(y)} \mathcal{A}_g^{(y)})}{\partial \epsilon} + \frac{\partial(F^{(x)} \mathcal{A}_g^{(x)})}{\partial \epsilon} + \frac{\partial(F^{(y)} \nabla_K \phi_d^{(y)})}{\partial \epsilon} \right] d\tau = \\ &\int_{t'}^t \left[F^{(y)} \frac{\partial \mathcal{A}_g^{(y)}}{\partial K_x} (K_x^{st} - A_x(\tau)) + (\mathcal{A}_g^{(x)} F^{(x)}) + F^{(y)} \frac{\partial \nabla_K \phi_d^{(y)}}{\partial K_x} (K_x^{st} - A_x(\tau)) \right] d\tau \end{aligned} \quad (6.9)$$

Along $\Gamma - K$, the C_2 symmetry dictates two conditions: $\mathcal{A}_g^{(x)} = 0$ and $\frac{\partial \mathcal{A}_g^{(y)}}{\partial K_x} = \frac{\partial \nabla_K \phi_d^{(y)}}{\partial K_x} = 0$. Therefore, along this axis, all first order ϵ -dependent terms vanish. However, along $\Gamma - M$ these three terms contribute to the *first order* perturbation. This perturbation is captured by the additional Berry phase, defined in the main text as $\Delta \gamma_B$ ($\Delta \gamma_B = \frac{\partial \gamma_B^{(i)}}{\partial \epsilon} \epsilon$). Note that this phase is accumulated along the entire trajectory and therefore is dominated by the time integrated terms $\tilde{\gamma}_B$.

Along the $\Gamma - M$ axis, we can write the ellipticity dependence of the odd and the

even harmonics intensities, using equation 6.5, as:

$$I^{(N)} \propto \begin{cases} C e^{-2Im(\Delta S_{inv})} (|d^y|^2 |\cos(\Delta\gamma_B(\epsilon))|^2 + |d^x|^2 |\sin(\Delta\gamma_B(\epsilon))|^2) & N \text{ is odd} \\ C e^{-2Im(\Delta S_{inv})} (|d^y|^2 |\sin(\Delta\gamma_B(\epsilon))|^2 + |d^x|^2 |\cos(\Delta\gamma_B(\epsilon))|^2) & N \text{ is even} \end{cases} \quad (6.10)$$

Where C represents all unperturbed prefactors.

6.2.1 Reconstruction of the Berry Phase

In the following section we describe the reconstruction procedure of the Berry phase accumulated along the trajectory, shaped by the elliptical field along $\Gamma - M$ (equation (6.10)). The signal of the odd and even harmonics provide two different measurements of the interferometer. In the first stage we combine neighboring odd and even harmonics according to:

$$I_{\Gamma-M}^+(\epsilon) = \frac{I_{\Gamma-M}^{odd}(\epsilon) + I_{\Gamma-M}^{even}(\epsilon)}{I_{\Gamma-M}^{odd}(0)} = f(\epsilon) (1 + r) \cosh(2Im(\Delta\gamma_B(\epsilon))) \quad (6.11)$$

$$I_{\Gamma-M}^-(\epsilon) = \frac{I_{\Gamma-M}^{odd}(\epsilon) - I_{\Gamma-M}^{even}(\epsilon)}{I_{\Gamma-M}^{odd}(0)} = f(\epsilon) (1 - r) \cos(2Re(\Delta\gamma_B(\epsilon))) \quad (6.12)$$

where $r = \frac{|d_x|^2}{|d_y|^2}$ is the dipole polarization ratio, and $f(\epsilon) = e^{-2Im(\Delta S_{inv}(\epsilon))}$ is the symmetric ellipticity coefficient. This coefficient represents the suppression of the harmonic signal with the ellipticity of the driving field. Here we assume that the dipoles, their amplitudes and phases, are not modified by the elliptical perturbation. We determine their values by the measurement at $\epsilon = 0$. Under these conditions, the dipole polarization ratio is the ratio between odd and even intensities (equation (6.6)),

$$r = \frac{I_{\Gamma-M}^{even}(0)}{I_{\Gamma-M}^{odd}(0)}.$$

We extract $f(\epsilon)$ by resolving the ellipticity response along $\Gamma - K$. The ellipticity response along this axis can be expressed as:

$$I_{\Gamma-K}^+(\epsilon) = \frac{I_{\Gamma-K}^{odd}(\epsilon) + I_{\Gamma-K}^{even}(\epsilon)}{I_{\Gamma-K}^{odd}(0) + I_{\Gamma-K}^{even}(0)} = f_{\Gamma-K}(\epsilon) \quad (6.13)$$

We note that this coefficient can be different along the two symmetry axes, $\Gamma - K$ and $\Gamma - M$, since it depends on the band dispersion. We correct for this difference using the two-color measurements (see next chapter for further details). Our experimental results show that the signal decay with ellipticity along $\Gamma - M$ is broader by 30% compared to $\Gamma - K$. Therefore, we consider such difference by taking: $f_{\Gamma-M} = 0.7 \cdot f_{\Gamma-K}$.

6.2.2 Theoretical Berry phase calculation

We perform a theoretical calculation of the interband Berry phase accumulated by the electron-hole wavepacket during a half cycle along the $\Gamma - M$ direction. We start with a density-functional theory (DFT) calculation of the α -quartz band structure and extract Wannier functions. From the the Wannier function-based Hamiltonian, we derive the Berry curvature and Berry connection. We focus on the band gap between an inner valence band (such to avoid level crossings which affect the numerics) and lowest conduction band (10.7 – 12.1 eV). In the next step we perform a semi-classical analysis, which calculates the k-space trajectory for each ionization time, for various values of the field's ellipticity. The calculations are performed for field intensity of 0.007 [a.u], wavelength of $\lambda = 1.2 [\mu m]$, for 21 different ionization times. We then numerically calculate the interband Berry phase acquired by the electron-hole wavepacket using the k-space trajectory, according to the following equation:

$$\gamma_{B,inter}(t', \epsilon) = - \int_{\mathbf{k}_{t'}^\epsilon(t')}^{\mathbf{k}_{t'}^\epsilon(t)} \mathcal{A}_g(\mathbf{k}) \cdot d\mathbf{k} + \phi_d^y(\mathbf{k}_{t'}^\epsilon(t)) - \phi_d^y(\mathbf{k}_{t'}^\epsilon(t')) \quad (6.14)$$

Where $\mathcal{A}_g(\mathbf{k}) = \mathcal{A}_c(\mathbf{k}) - \mathcal{A}_v(\mathbf{k})$ is the conduction-valence Berry connection difference, and ϕ_d^y is the conduction-valence dipole phase along the ionization direction. Equation (6.14) provides the gauge invariant loop through crystal momentum-energy space that introduces the interband Berry phase. In this scheme an electron is ionized at t' , accumulating the ionization dipole phase $\phi_d^y(\mathbf{k}_{t'}^\epsilon(t'))$, then the electron and hole propagate, each one in their band, accumulating the Berry connection at each k-point, until finally at time t they recombine, accumulating the recombination dipole phase $\phi_d^y(\mathbf{k}_{t'}^\epsilon(t))$ and closing the loop in parameter space.

Once we calculate the interband Berry phase for each ionization time, we map

the recombination energies of each trajectory to its emission energy. We consider the finite bandwidth of each harmonic by averaging the phase over ionization times associated with an energy window of ± 0.1 [eV]. We perform this procedure for various values of the field’s ellipticity. Finally, in the experiment we are sensitive to the anti-symmetric part of the interband Berry phase, only. Therefore, we anti-symmetrise our results, and take their absolute value.

Since our calculation is limited to the first band gap, we focus on studying the Berry phase dependence on the recombination energy (can be mapped to the photon energy in the HHG spectrum). Here each recombination energy represents a different trajectory in momentum-energy space. Using this calculation we can visualize how the accumulated berry phase follows the wavepacket k-space evolution.

Figure 14 shows the accumulated interband Berry phase for the roughly shortest trajectory (corresponds to 11eV recombination energy) and the longest trajectory (12eV). As can be seen by our calculation, as the trajectory length increases– the accumulated Berry phase is larger. This validates our reconstruction results where the reconstructed Berry phase increases with harmonic order (figure 2 in the main text). Finally, the calculated Berry phase at high ellipticity values ranges between $0.8 - 1.2$ [rad] which is close to the reconstructed Berry phase of harmonics 14-15. This is due to the similar trajectory length associated with these these harmonics.

6.3 Two-color Field

We observe the role of the Berry curvature by applying a driving field with a mirror symmetry between the two consecutive half cycles. In this case, the induced transverse perturbation is symmetric (figure 3b in the main text): $(k_x, k_y) \rightarrow (k_x, -k_y)$. Therefore, in strong contrast to the elliptically polarized field case (chapter 6.2), the two color field does not induce a phase difference between the two interfering trajectories, associated with the Berry phase. Following equation (6.9), one can see that

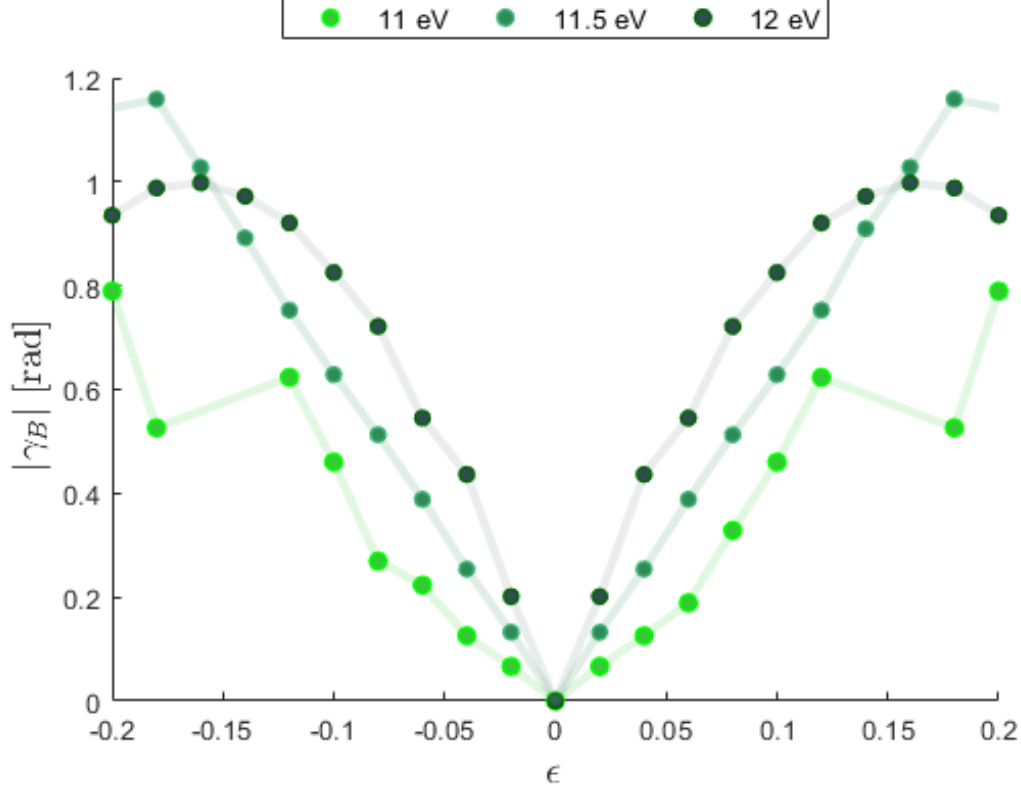


Figure 14: Interband Berry phase for trajectories associated with different recombination energies.

the first order perturbation is symmetric with respect to the two half-cycles:

$$\begin{aligned}
 [F^{(y)}(t) \frac{\partial \mathcal{A}_g^{(y)}}{\partial K_x}]_{K_x=0, K_y} (K_x - A_x(\tau)) &= [F^{(y)}(t + \frac{T}{2}) \frac{\partial \mathcal{A}_g^{(y)}}{\partial K_x}]_{K_x=0, -K_y} (K_x - A_x(\tau)) = \\
 &(-1)^2 F^{(y)}(t) \frac{\partial \mathcal{A}_g^{(y)}}{\partial K_x} \Big|_{K_x=0, K_y} (K_x - A_x(\tau)) \\
 [\mathcal{A}_g^{(x)}]_{K_x=0, K_y} F^{(x)}(t) &= [\mathcal{A}_g^{(x)}]_{K_x=0, -K_y} F^{(x)}(t + \frac{T}{2})
 \end{aligned} \tag{6.15}$$

Our experimental results confirm the absence of a phase difference, showing a similar response of the odd and the even harmonics, with respect to the two color

delay. However, due to the symmetry of the driving field, the effect of the Berry curvature on the wavepackets evolution is not canceled out and dominates the harmonic signal.

Using the semi-classical equation, we can write the evolution of the electron-hole wavepacket in real space and extract their recombination condition (chapter 4). For linearly polarized driving field (y direction), along the main axes of the crystal, the 2D wavepacket evolution is described by:

$$\begin{aligned}
\dot{k}_y &= -A_0^y \sin(\omega_0 t) \\
\dot{k}_x &= 0 \\
\Delta y(t', t) &= \int_{t'}^t \frac{\partial \varepsilon_g(k_x, k_y(\tau))}{\partial k_y} d\tau \\
\Delta x(t', t) &= - \int_{t'}^t F_0^y \cos(\omega_0 \tau) \Omega_g^z(k_x, k_y(\tau)) d\tau
\end{aligned} \tag{6.16}$$

Consider t' as the ionization time, the relative displacement of the hole and the electron at a given time t is $\Delta x, \Delta y$. The recombination condition, which dictates the recombination time t is $\Delta x = \Delta y = 0$.

According to Time Reversal Symmetry, $\varepsilon(-\mathbf{k}) = \varepsilon(\mathbf{k})$, $\mathbf{\Omega}(-\mathbf{k}) = -\mathbf{\Omega}(\mathbf{k})$, the evolution along the y direction is reversed during the two subcycles of the driving field, while along the x direction it remains the same. Therefore, in the presence of a Berry curvature, the wavepackets evolution along the two half cycles are mirror image of each other (as presented in figure 3 in the main text and in chapter 4.2).

Adding a weak second harmonic field (SH), orthogonally polarized in respect to the fundamental field, $\epsilon A_0^y \sin(2\omega_0 t + \varphi)$, induces an additional evolution along the transverse direction:

$$\begin{aligned}
\dot{k}_x &= -\epsilon A_0^y \sin(2\omega_0 t + \varphi) \\
\Delta x(t', t, \varphi) &= \int_{t'}^t \frac{\partial \varepsilon_g(k_x(\tau, \varphi), k_y(\tau))}{\partial k_x} - F_0^y \cos(\omega_0 \tau) \Omega_g^z(k_x(\tau, \varphi), k_y(\tau)) d\tau
\end{aligned} \tag{6.17}$$

The SH field contribution to the transverse displacement depends on φ , the delay between the two fields. Therefore, by scanning this delay, we can compensate

or enhance the transverse displacement caused by the original anomalous velocity. Since the HHG signal is extremely sensitive to the recombination probability, this manipulation is mapped directly to the harmonic intensities [22]. Note that here we assume that due to the perturbative nature of the second harmonic field, the ionization time, recombination times and the y direction propagation are not modified, being dictated by the fundamental field only.

In order to take a deeper insight, we study the dependence of the harmonic signal on the two color delay. As was previously studied in gas phase HHG spectroscopy, the harmonic intensity depends exponentially on the recombination displacement, $\exp^{-C|\Delta x|^2}$ (where C is a constant), being insensitive to its sign [23]. Therefore, in absence of the anomalous velocity, where the Berry curvature is zero, the harmonics signal oscillates at an even integer of the SH frequency:

$$\begin{aligned} |\Delta x(\varphi)|^2 &= \int_{t'}^t \frac{\partial \varepsilon_g(k_x(\tau, \varphi), k_y(\tau))}{\partial k_x} d\tau \\ &= \left| \int_{t'}^t (a_1 k_x(\tau, \varphi) + a_3 (k_x(\tau, \varphi))^3 + \dots) d\tau \right|^2 \propto b_2 \sin(2\varphi + \varphi_2) + b_4 \sin(4\varphi + \varphi_4) \dots \end{aligned} \quad (6.18)$$

Where $a_1, a_3, \dots$, $b_2, b_4, \dots$ and $\varphi_2, \varphi_4, \dots$ are constants. However, along crystal orientations associated with non-vanishing Berry curvature, the anomalous velocity contribution includes a term that is independent of the two color delay. This can be obtain by expanding the anomalous velocity contribution up to the first order in ϵ , $F_0^y \cos(\omega_0 \tau) [\frac{\partial \Omega_g}{\partial k_x}(k_x(\tau, \varphi)) + \Omega_g(k_y(\tau))]$, reducing the oscillation frequency of the harmonic signal:

$$\begin{aligned} |\Delta x(\varphi)|^2 &= \left| \int_{t'}^t (a_1 k_x(\tau, \varphi) + a_3 (k_x(\tau, \varphi))^3 + \dots + F_0^y \cos(\omega_0 \tau) [\Omega_g(k_y(\tau)) + \frac{\partial \Omega_g}{\partial k_x}(k_x(\tau, \varphi))]) d\tau \right|^2 \\ &\propto c_0 + c_1 \sin(\varphi + \varphi_1) + c_2 \sin(2\varphi + \varphi_2) \dots \end{aligned} \quad (6.19)$$

Where $c_0, c_1, \dots$ are constants and proportional to the Berry curvature and the field

amplitude $c_0 \propto F_0^y \Omega_g$. This simple picture is reflected in our experimental results, presented in figure 3 in the main text.

6.3.1 Theoretical study

To support the experimental findings in Fig.3c (see main text), we performed numerical calculations on a two-band gapped-graphene model [24], which captures the essential physics discussed in the manuscript. The most notable differences are that the minimum band gap for gapped graphene is at the K and K' points, as opposed to the Γ point in α quartz, and that the mirror symmetry direction is along Γ -M for gapped graphene, while it is along Γ -K for α quartz. The Berry curvature of gapped graphene is shown in Fig. 15a). We chose the minimum band gap of the model system equal to that reported for α quartz (9 eV), and used similar laser parameters to those estimated for the experiment: a linearly-polarized 1.2 μm and its perpendicular second harmonic, with intensities of 30 TW/cm² and 0.08 TW/cm², respectively.

We performed calculations for three different polarization angles with respect to the sample: 0° (Γ -M), 5° and 30°. For each polarization angle, we performed a time delay scan between the ω and 2ω pulses on the range $[0, \frac{2\pi}{2\omega}]$, and extracted the integrated harmonic yield for each delay. Fig. 15b-d show the oscillation of the harmonic yield as a function of the two-color time delay for the three different polarization angles.

When the laser is polarized along the mirror symmetry direction (Fig. 15b, to be compared with the 0° panel in Fig.3c), the harmonic yield beats with the characteristic 4ω periodicity of ω - 2ω experiments in inversion symmetric targets, as explained in the main text and as observed in Fig.3c. As the polarization direction is moved away from the mirror symmetry axis, the period reduces to one cycle of the 2ω field (Fig. 15c,d, to be compared with the 5° and 30° panels of Fig.3c, respectively), as explained in the main text.

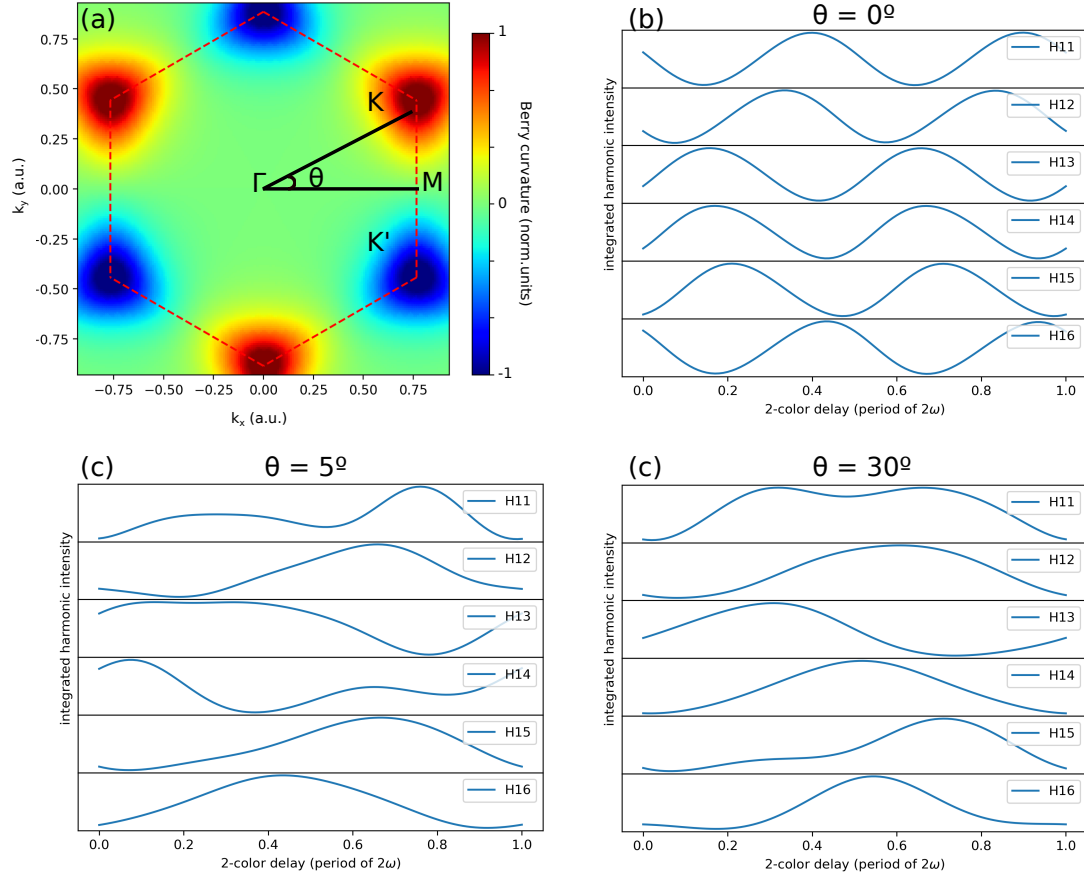


Figure 15: Two-color numerical simulations for a gapped-graphene model. (a) Berry curvature in the first Brillouin zone (red-dashed hexagon) of the conduction band. (b-d) Integrated yield of harmonics 11 to 16 as a function of the two-color time delay, for polarization directions along $\theta = 0^\circ$ (b), $\theta = 5^\circ$ (c) and $\theta = 30^\circ$ (d).

References

- [1] Georg Kresse and Jürgen Furthmüller. Efficiency of ab-initio total energy calculations for metals and semiconductors using a plane-wave basis set. *Computational materials science*, 6(1):15–50, 1996.
- [2] Georg Kresse and Jürgen Furthmüller. Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set. *Physical Review B*, 54(16):11169, 1996.
- [3] John P Perdew, Kieron Burke, and Matthias Ernzerhof. Generalized gradient approximation made simple. *Physical Review Letters*, 77(18):3865, 1996.
- [4] Peter E Blöchl. Projector augmented-wave method. *Physical Review B*, 50(24):17953, 1994.
- [5] Georg Kresse and Daniel Joubert. From ultrasoft pseudopotentials to the projector augmented-wave method. *Physical Review B*, 59(3):1758, 1999.
- [6] Giovanni Pizzi, Valerio Vitale, Ryotaro Arita, Stefan Blügel, Frank Freimuth, Guillaume Géranton, Marco Gibertini, Dominik Gresch, Charles Johnson, Takashi Koretsune, et al. Wannier90 as a community code: new features and applications. *Journal of Physics: Condensed Matter*, 32(16):165902, 2020.
- [7] Tran Trung Luu and Hans Jakob Wörner. Measurement of the Berry curvature of solids using high-harmonic spectroscopy. *Nat Commun*, 9(1):916, December 2018.
- [8] Hanzhe Liu, Yilei Li, Yong Sing You, Shambhu Ghimire, Tony F Heinz, and David A Reis. High-harmonic generation from an atomically thin semiconductor. *Nature Physics*, 13(3):262, 2017.
- [9] Shambhu Ghimire, Anthony D DiChiara, Emily Sistrunk, Pierre Agostini, Louis F DiMauro, and David A Reis. Observation of high-order harmonic generation in a bulk crystal. *Nature physics*, 7(2):138, 2011.

- [10] Alexis Chacón, Dasol Kim, Wei Zhu, Shane P. Kelly, Alexandre Dauphin, Emilio Pisanty, Andrew S. Maxwell, Antonio Picón, Marcelo F. Ciappina, Dong Eon Kim, Christopher Ticknor, Avadh Saxena, and Maciej Lewenstein. Circular dichroism in high-order harmonic generation: Heraldng topological phases and transitions in Chern insulators. *Phys. Rev. B*, 102(13):134115, October 2020. arXiv: 1807.01616.
- [11] G. Vampa, C. R. McDonald, G. Orlando, P. B. Corkum, and T. Brabec. Semi-classical analysis of high harmonic generation in bulk crystals. *Physical Review B*, 91(6):064302, February 2015.
- [12] Ayelet Julie Uzan, Gal Orenstein, Álvaro Jiménez-Galán, Chris McDonald, Rui E. F. Silva, Barry D. Bruner, Nikolai D. Klimkin, Valerie Blanchet, Talya Arusi-Parpar, Michael Krüger, Alexey N. Rubtsov, Olga Smirnova, Misha Ivanov, Binghai Yan, Thomas Brabec, and Nirit Dudovich. Attosecond spectral singularities in solid-state high-harmonic generation. *Nature Photonics*, 14(3):183–187, March 2020. Bandiera_abtest: a Cg_type: Nature Research Journals Number: 3 Primary_atype: Research Publisher: Nature Publishing Group Subject_term: Attosecond science;Condensed-matter physics Subject_term_id: attosecond-science;condensed-matter-physics.
- [13] Giulio Vampa, TJ Hammond, Nicolas Thiré, BE Schmidt, François Légaré, CR McDonald, Thomas Brabec, DD Klug, and PB Corkum. All-optical reconstruction of crystal band structure. *Physical review letters*, 115(19):193603, 2015.
- [14] G Vampa, CR McDonald, G Orlando, PB Corkum, and T Brabec. Semiclassical analysis of high harmonic generation in bulk crystals. *Physical Review B*, 91(6):064302, 2015.
- [15] G Vampa, CR McDonald, G Orlando, DD Klug, PB Corkum, and T Brabec. Theoretical analysis of high-harmonic generation in solids. *Physical review letters*, 113(7):073901, 2014.

- [16] G Vampa, TJ Hammond, N Thiré, BE Schmidt, F Légaré, CR McDonald, T Brabec, and PB Corkum. Linking high harmonics from gases and solids. *Nature*, 522(7557):462, 2015.
- [17] Ayelet Julie Uzan, Gal Orenstein, Álvaro Jiménez-Galán, Chris McDonald, Rui EF Silva, Barry D Bruner, Nikolai D Klimkin, Valerie Blanchet, Talya Arusi-Parpar, Michael Krüger, et al. Attosecond spectral singularities in solid-state high-harmonic generation. *Nature Photonics*, 14(3):183–187, 2020.
- [18] Lun Yue and Mette B. Gaarde. Imperfect Recollisions in High-Harmonic Generation in Solids. *Phys. Rev. Lett.*, 124(15):153204, April 2020.
- [19] János K Asbóth, László Oroszlány, and András Pályi. A short course on topological insulators. *Lecture notes in physics*, 919:166, 2016.
- [20] Shicheng Jiang, Hui Wei, Jigen Chen, Chao Yu, Ruifeng Lu, and C. D. Lin. Effect of transition dipole phase on high-order-harmonic generation in solid materials. *Phys. Rev. A*, 96(5):053850, November 2017.
- [21] Shicheng Jiang, Chao Yu, Jigen Chen, Yanwei Huang, R. F. Lu, and C. D. Lin. How to obtain complex transition dipole moments satisfying crystal symmetry and periodicity from ab-initio calculations. *Phys. Rev. B*, 102(15):155201, October 2020. arXiv: 1912.09649.
- [22] Dror Shafir, Hadas Soifer, Barry D Bruner, Michal Dagan, Yann Mairesse, Serguei Patchkovskii, Misha Yu Ivanov, Olga Smirnova, and Nirit Dudovich. Resolving the time when an electron exits a tunnelling barrier. *Nature*, 485(7398):343, 2012.
- [23] Yann Mairesse, Nirit Dudovich, J Levesque, M Yu Ivanov, Paul B Corkum, and DM Villeneuve. Electron wavepacket control with elliptically polarized laser light in high harmonic generation from aligned molecules. *New Journal of physics*, 10(2):025015, 2008.

- [24] Álvaro Jiménez-Galán, Rui EF Silva, Olga Smirnova, and Misha Ivanov. Sub-cycle valleytronics: control of valley polarization using few-cycle linearly polarized pulses. *Optica*, 8(3):277–280, 2021.