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Characterizing Anomalous High-Harmonic Generation in Solids

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Anomalous high-harmonic generation (HHG) arises in certain solids when irradiated by an intense laser field, originating from a Berry-curvature-induced perpendicular anomalous current. The observation of pure anomalous harmonics is, however, often prohibited by contamination from harmonics stemming from interband coherences. Here, we fully characterize the anomalous HHG mechanism, via development of an ab-initio methodology for strong-field laser-solid interaction that allows a rigorous decomposition of the total current. We identify two unique properties of the anomalous harmonic yields: an overall yield increase with laser wavelength; and pronounced minima at certain laser wavelengths and laser intensities around which the spectral phases drastically change. Such signatures can be exploited to disentangle the anomalous harmonics from competing HHG mechanisms, and thus pave way for the experimental identification and time-domain control of pure anomalous harmonics, as well as reconstruction of Berry curvatures.

When a charge carrier in a crystal with a nonvanishing Berry curvature is perturbed by a weak electric field, it acquires an anomalous perpendicular velocity component [1–5] and a corresponding anomalous current. Such anomalous velocities and Berry curvatures are ubiquitous in modern physics and are e.g. responsible for various Hall effects [6, 7]. With the advancements in light-source technologies [8, 9] and the availability of intense laser pulses, it is now possible to induce highly nonperturbative anomalous currents that are sources for anomalous high-harmonic generation (HHG) [10–14]. The broadening of our understanding of the anomalous current from the conventional low-energy condensed-matter physics to the extreme nonperturbative regime [15–18] is of utmost importance for both fundamental and application purposes.

In the strong-field regime, it is a challenge to extract detailed information from experiment and theory, given the multitude of nonperturbative phenomena that are driven simultaneously in a material. Such a situation is sketched in Fig. 1, in which a linearly polarized laser field induces two perpendicular currents: an anomalous current and an interband current [19]. An open question is which of these contributions dominate, a subject that has been under contention in recent interpretations of HHG experiments: the measured perpendicular-polarized harmonics have been attributed entirely to either Berry curvatures [10–12, 14] or interband coherences [20–24]. Similarly, no theoretical studies on the nonperturbative anomalous current have fully characterized its competition with the other generation mechanisms [25–27]. A theoretical framework that can facilitate the rigorous isolation of the anomalous HHG, and provide signatures and guidelines for their experimental identification is highly desired.

The generation and identification of pure anomalous currents and high harmonics are, besides the fundamental aspects, also relevant for application and spectroscopy purposes. It is well-known that different generation

mechanisms exhibit distinct characteristics in the spectral and temporal domains, where e.g. distinct sub-cycle emission features had previously aided experiments in determining the underlying HHG mechanisms in quartz as intraband [28, 29], and in ZnO as interband [30, 31]. Determining similar characteristics for anomalous HHG would aid in the understanding of phase-matching conditions for the development of new light sources [16, 29, 32], as well as the optical control of electronic currents at the petahertz rates [28, 33–36]. Additionally, generating pure anomalous harmonics is a prerequisite for Berry-curvature high-harmonic spectroscopy [11, 14], which adds to the already burgeoning field of high-harmonic spectroscopy [12, 13, 28–31, 37–49].

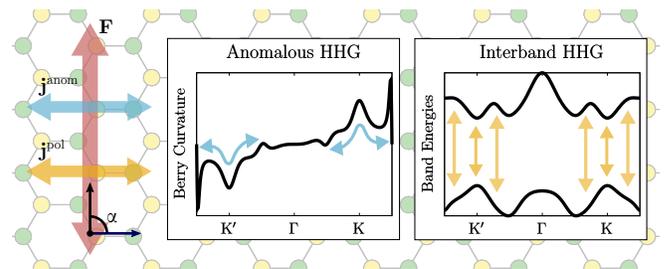


FIG. 1. In a crystal, a strong laser field $\mathbf{F}(t)$, polarized at an angle α relative to the mirror axis of a hexagonal crystal, induces nonperturbative perpendicular currents $\mathbf{j}^{\text{anom}}(t)$ and $\mathbf{j}^{\text{pol}}(t)$ that are sources for anomalous and interband HHG. The insets sketch the underlying generation mechanisms.

In this letter, we characterize the unique properties of the anomalous HHG current, allowing us to distinguish it from the other contributions through its intensity- and wavelength dependence as well as its subcycle emission dynamics. For this purpose, we develop an ab-initio methodology for strong-field light-matter interaction that allows a rigorous decomposition of the total current into its Bloch-gauge-invariant constituents, without the explicit need for the construction of a globally smooth

and periodic Bloch gauge [7, 50–55]. We consider monolayer MoS₂ as our specific target system for HHG, due to its potential technological applications [56–58] and the absence of propagation effects. We identify two main signatures of the anomalous harmonics: (i) a yield increase with laser wavelength and dominance over the interband harmonics below a threshold harmonic energy; (ii) characteristic minima in the yield for certain laser wavelengths or intensities around which the sub-cycle time structure drastically changes. We employ a semiclassical model to extract the underlying physics, whereby signature (ii) is traced to a π -jump of the harmonic spectral phase. Our results add to the fundamental understanding of nonperturbative anomalous currents, provide a guide towards experimental identification and control of pure anomalous harmonics, and resolve an open debate on the origin of perpendicular-polarized high harmonics in solids [10–12, 14, 20–24].

Our ab-initio methodology starts with the calculation of the monolayer MoS₂ band structure $E_n^{\mathbf{k}}$ and momentum coupling matrix elements $\mathbf{p}_{mn}^{\mathbf{k}}$, employing density functional theory [59] with ONCV pseudopotentials [60, 61] and PBE functionals [62]. Atomic units are used throughout this work unless indicated otherwise. The Brillouin zone is sampled with $N = 8100$ points in a Monkhorst-Pack mesh. For the dynamics, we solve the time-dependent equations for the density matrix elements $\rho_{mn}^{\mathbf{k}}(t)$ in the velocity gauge [53, 55, 63], taking into account 90 bands. At every 5th step of the time propagation, we transform into an adiabatic basis [55] to include a dephasing time $T_2 = 10$ fs, and decompose the total current into four Bloch-gauge-invariant terms, $\mathbf{j}(t) = \mathbf{j}^{\text{tra}}(t) + \mathbf{j}^{\text{pol}}(t) + \mathbf{j}^{\text{anom}}(t) + \mathbf{j}^{\text{mix}}(t)$, where

$$\mathbf{j}^{\text{tra}}(t) = -N^{-1} \sum_{n\mathbf{k}} \tilde{\mathbf{p}}_{nn}^{\mathbf{k}}(t) \tilde{\rho}_{nn}^{\mathbf{k}}(t), \quad (1a)$$

$$\mathbf{j}^{\text{pol}}(t) = -N^{-1} \partial_t \sum_{m \neq n, \mathbf{k}} \tilde{\mathbf{d}}_{mn}^{\mathbf{k}}(t) \tilde{\rho}_{nm}^{\mathbf{k}}(t), \quad (1b)$$

$$\mathbf{j}^{\text{anom}}(t) = -N^{-1} \sum_{n\mathbf{k}} \left[\mathbf{F}(t) \times \tilde{\mathbf{\Omega}}_n^{\mathbf{k}}(t) \right] \tilde{\rho}_{nn}^{\mathbf{k}}(t), \quad (1c)$$

$$\mathbf{j}^{\text{mix}}(t) = -N^{-1} \sum_{\mu=x,y} F_{\mu}(t) \sum_{m \neq n, \mathbf{k}} [\tilde{d}_{\mu,mn}^{\mathbf{k}}(t)]_{;\mathbf{k}} \tilde{\rho}_{nm}^{\mathbf{k}}(t), \quad (1d)$$

with $\mathbf{j}^{\text{tra}}(t)$ the intraband current, $\mathbf{j}^{\text{pol}}(t)$ the interband (polarization) current, $\mathbf{j}^{\text{anom}}(t)$ the anomalous current, and $\mathbf{j}^{\text{mix}}(t)$ the mixture current originating in the coupling between the intraband and interband position operators [27, 55, 64, 65]. The tildes in Eq. (1) indicate quantities evaluated in the adiabatic basis. After calculation of the current decomposition at time t , we transform back into the Bloch basis and continue with the time-dependent propagation of $\rho_{mn}^{\mathbf{k}}(t)$. We evaluate the dipole matrix elements $\tilde{\mathbf{d}}_{mn}^{\mathbf{k}}(t)$, generalized gradients $[\tilde{d}_{\mu,mn}^{\mathbf{k}}(t)]_{;\mathbf{k}}$ [65, 66], and Berry curvatures $\tilde{\mathbf{\Omega}}_n^{\mathbf{k}}(t)$

entirely in terms of $\tilde{E}_n^{\mathbf{k}}(t)$ and $\tilde{\mathbf{p}}_{mn}^{\mathbf{k}}(t)$ by using relevant sum-rules [65–67]. The key advantage of this methodology is that it does not require the construction of a globally smooth and periodic gauge for the Bloch states, in contrast to previous methods that decompose the current [27, 65, 68], but instead requires more bands for convergence (see Supplemental Material (SM) [63]). The electric field, $\mathbf{F}(t) = -\partial_t \mathbf{A}(t)$, is linearly polarized in the plane of the monolayer, and the vector potential $\mathbf{A}(t)$ has a total duration of 12 optical cycles and a \cos^2 -envelope. Since $\mathbf{j}^{\text{anom}}(t) \perp \mathbf{F}(t)$, we focus our attention on the harmonic spectra polarized perpendicular to the laser field, $S(\omega) = \omega^2 |\mathbf{j}(\omega) \cdot \hat{\mathbf{e}}_{\perp}|^2$, with $\mathbf{j}(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \mathbf{j}(t)$. **Note that excitonic effects have been neglected in this work. Although this is appropriate for the system/regime we are in according to [15, 63], it is an ongoing effort in the community to understand excitonic effects on HHG [69, 70].**

We highlight that we do not label the anomalous current as an intraband contribution: even though $\mathbf{j}^{\text{anom}}(t)$ in Eq. (1c) does not depend on the time-dependent coherences, the Berry curvature for the n th band arises from the residual coupling between the other bands [6, 63, 67]. In addition, it can be easily shown that the oft-used expression for the “interband” current $N^{-1} \sum_{m \neq n, \mathbf{k}} \tilde{\mathbf{p}}_{mn}^{\mathbf{k}}(t) \tilde{\rho}_{nm}^{\mathbf{k}}(t)$ equals $\mathbf{j}^{\text{pol}}(t) + \mathbf{j}^{\text{anom}}(t) + \mathbf{j}^{\text{mix}}(t)$. In this sense, the experimentally measured perpendicular harmonics interpreted as “interband” [20–24] likely already contains the anomalous contribution, while in this work interband current refers to $\mathbf{j}^{\text{pol}}(t)$ of Eq. (1b) [71]. Considering $\mathbf{j}^{\text{anom}}(t)$ as a stand-alone contribution to the total current facilitates a link between the weak-field condensed matter community, where Berry curvatures and anomalous velocities are ubiquitous, and the strong-field community. As we will see, the anomalous high harmonics exhibit unique characteristics separating them from other HHG contributions.

Figure 2(a) shows the decomposed perpendicular-polarized harmonic spectrum for a field with carrier wavelength 7.4 μm , intensity 50 GW/cm² and $\alpha = 30^\circ$ (α defined in Fig. 1). Only even-order harmonics are observed, with the interband and anomalous contributions clearly competing: the interband harmonics dominate at higher orders, even well below the band gap energy at around harmonic 8 (H8) and H10; while the anomalous harmonics dominate for the lowest orders at H2-H6. The intraband and mixture currents contribute with yields that are order(s) of magnitude lower.

Figures 2(b) and 2(c) show respectively the anomalous and interband HHG spectra versus α . Our results conform with the dynamical-symmetry selection rules for HHG [72], where the D_{3h} spatial point group of monolayer MoS₂ together with the in-plane linearly-polarized field prohibit the emission of all perpendicular harmonics at $\alpha = n60^\circ$ and odd-order harmonics at $\alpha = 30^\circ + n60^\circ$, with n an integer [10, 72–74]. The anomalous harmonics

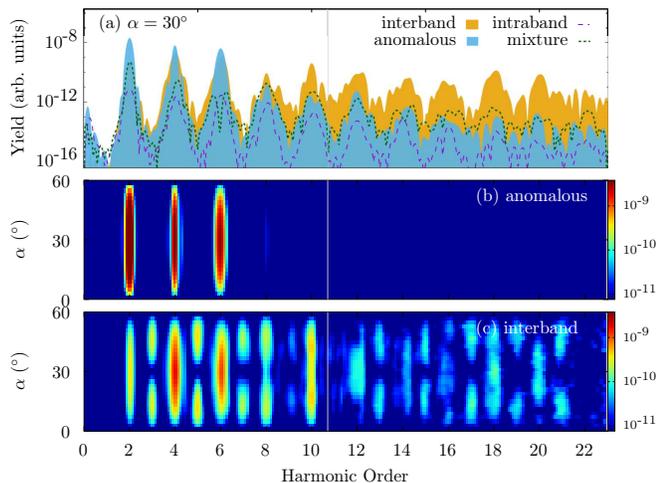


FIG. 2. Spectra for perpendicular-polarized harmonics in monolayer MoS₂ for a 7.4 μm driver. (a) Different current-contributions to spectrum for $\alpha = 30^\circ$. The (b) anomalous and (c) interband harmonics versus α . The gray vertical lines mark the gap energy.

in Fig. 2(b) only permit even orders for all α , since time-reversal symmetry dictates $\Omega_m^{-\mathbf{k}} = -\Omega_m^{\mathbf{k}}$ which leads to $\mathbf{j}^{\text{anom}}(t + \frac{T}{2}) = \mathbf{j}^{\text{anom}}(t)$, with T the optical period. In Fig. 2(c), no symmetry rule prohibits the emission of interband harmonics between $\alpha = 0^\circ$ and $\alpha = 30^\circ$, and both even and odd orders are observed.

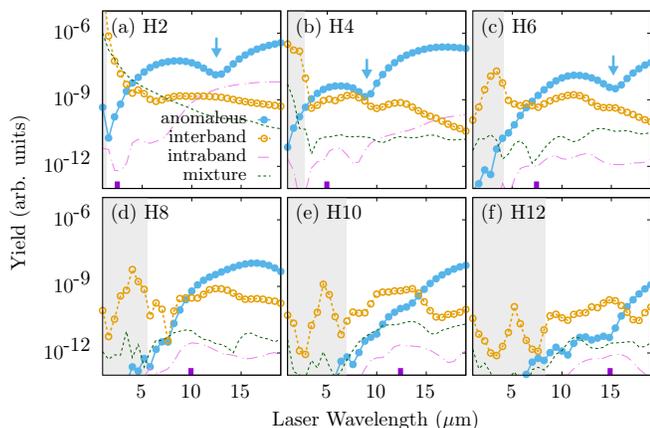


FIG. 3. Decomposed harmonic yields versus laser wavelength for $\alpha = 30^\circ$. The arrows in (a)-(c) mark pronounced minima in the anomalous harmonic yield. In each panel, the gray shaded areas indicate that the harmonic energy is larger than the band gap. Purple ticks on the wavelength axis mark the harmonic energy 1 eV.

To further characterize the anomalous HHG mechanism and its competition with the other generation mechanisms, we show in Fig. 3 the decomposed harmonic yields for H2-H12 versus laser wavelength, fixing the laser intensity at 50 GW/cm² and $\alpha = 30^\circ$. We focus on the anomalous and interband contributions, which

are seen to dominate over the intraband and mixture contributions for almost all wavelengths. At shorter laser wavelengths, in the gray shaded regions of Fig. 3 where the harmonic energy is greater than the gap energy of 1.79 eV, the interband yield dominates over the anomalous yield for all harmonic orders. With increasing wavelength, the anomalous harmonic yield overall increases, and begins to dominate over the interband yield at around a harmonic energy of $E_h = 1$ eV (marked by purple ticks on the wavelength axis). At the longest wavelength in the figure, 19 μm , the anomalous harmonics dominate completely – e.g. the anomalous H6 yield at 19 μm is around 3 orders of magnitudes greater than the interband yield. We judge the transition energy E_h to be material dependent and governed by the particular band structure and coupling matrix elements of the system, but nonetheless expect it to be generally smaller than the gap energy, as above-band-gap harmonics generally proceeds via interband coherences [71, 75]. Figure 3 clears up the recent contention on the origin of the perpendicular harmonics in MoS₂ in terms of either anomalous [10, 76, 77] or interband harmonics [21–24, 76, 77]: these two harmonic contributions dominate in different laser-parameter and harmonic-energy regimes.

In Figs. 3(a)-3(c), the anomalous harmonic yield for H2, H4 and H6 exhibit prominent minima marked by blue arrows at 12.5 μm , 9 μm and 15.2 μm , respectively. We performed additional HHG simulations using a fixed laser wavelength of 7.4 μm and varying laser intensities, and observed similar pronounced minima at specific intensities (see SM [63]). These minima represent a striking characteristic of the anomalous harmonics, which we now characterize employing a semiclassical model.

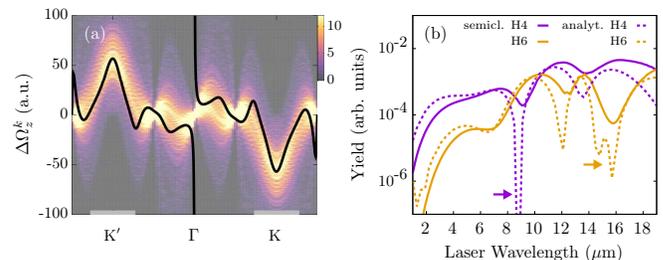


FIG. 4. Berry-curvatures: calculated from first principles (solid line); reconstructed from the HHG spectrum in Fig. 2(b) (color plot), with the gray lines marking the excursion regions of electrons and holes created at K and K'. (b) Semiclassical numerical and analytical results (see text) for the anomalous H4 and H6 yields versus wavelength.

We write the semiclassical anomalous current as $\mathbf{j}^{\text{SC}}(t) \propto -\mathbf{F}(t) \times [\Delta\Omega^{\mathbf{K}+\mathbf{A}(t)} + \Delta\Omega^{\mathbf{K}'+\mathbf{A}(t)}]$, in which we assume that the electrons tunnel at the minimum band gaps at K and K' [10, 78], and adopt a two-band approximation, such that $\Delta\Omega^{\mathbf{k}}$ is the Berry curvature difference between the lowest CB and the highest VB, representing

the total contribution from the electron and hole. The solid line in Fig. 4(a) shows the z -component of $\Delta\Omega^{\mathbf{k}}$ along $K'-\Gamma-K$. The calculated semiclassical yields, using the same laser fields as in the ab-initio calculations of Fig. 3, are plotted in Fig. 4(b) for H4 and H6 in solid lines. Clear minima are observed, with the most pronounced minimum seen at $\sim 9 \mu\text{m}$ for H4 and at $\sim 15.7 \mu\text{m}$ for H6, consistent with our ab-initio results in Fig. 3. To gain further insight, we assume a monochromatic vector potential $\mathbf{A}(t) = F_0/\omega_0 \sin(\omega_0 t)\hat{\mathbf{e}}$, which leads to the analytical expression for the current, $\mathbf{j}^{\text{SC}}(t) \propto \sum_{s=1}^{\infty} C_{2s} \sin(2s\omega_0 t)$, with

$$C_{2s} = \frac{s\omega_0}{a} \sum_{q=1}^{\infty} \frac{\Omega_q}{q} \cos\left(\frac{2q\pi}{3}\right) J_{2s}\left(\frac{qaF_0}{2\omega_0}\right) \quad (2)$$

the amplitudes, Ω_q the Fourier coefficients of $\Delta\Omega_z^{\mathbf{k}}$, J_{2s} the $(2s)$ th-order Bessel function, and a the lattice constant. The yield versus wavelength for the $(2s)$ th anomalous harmonic, $S_{2s} \propto s^2\omega_0^2 |C_{2s}|^2$, is plotted in Fig. 4(b) for $2s = 4$ and $2s = 6$ in dashed lines. The pronounced minima marked by the arrows hence correspond to nodes of C_{2s} seen as a function of wavelength. We see that the expression for C_{2s} in Eq. (2) involves both the nonlinearities of the Bessel functions and the Berry curvature Fourier coefficients Ω_q . We have checked that the q -sum in Eq. (2) is not dominated by a single term, but rather a superposition of terms. In addition, the pronounced minima in the yield-versus-laser-intensity plots from the ab-initio calculations are also explained in terms of the nodes of C_{2s} (considered as a function of the laser intensity) [63]. The nodes in C_{2s} is another characterization tool for the anomalous harmonics, as the presence of such minima in a wavelength or intensity scan is an indication of the dominance of the anomalous HHG mechanism.

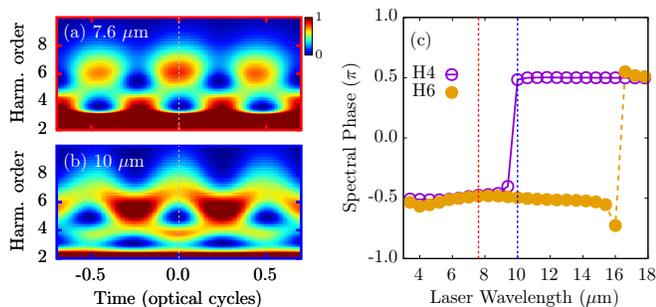


FIG. 5. Time-frequency emission profiles for the anomalous harmonics extracted from the ab-initio simulations for laser wavelength (a) $7.6 \mu\text{m}$ and (b) $10 \mu\text{m}$. The colorbar axis is in linear scale and arbitrary units, and the dashed vertical line is to guide the eyes. (c) Spectral phases for the anomalous H4 and H6 extracted from the simulations, with the wavelengths used in (a) and (b) marked by vertical lines.

Finally, we consider the emission-time profiles and the spectral phases of the solid-state harmonics, which en-

code pertinent information about the underlying harmonic emission mechanisms [28–31]. In Figs. 5(a) and 5(b), we show the time-frequency profiles for the anomalous harmonic emissions obtained from the ab-initio calculations, at laser wavelengths corresponding to before and after the H4 minimum at $\lambda_{min} \equiv 9 \mu\text{m}$ [Fig. 3(b)], respectively. For the $7.6 \mu\text{m}$ case in Fig. 5(a), the spectral yields around H6 is emitted at $t = T/2$, corresponding to the electric field extrema, while H4 is emitted at $t = (n + \frac{1}{2})T/2$ at the zeros of the field. For the longer laser wavelength $10 \mu\text{m} > \lambda_{min}$ in Fig. 5(b), the emission time of H4 and H6 are shifted by a quarter optical cycle.

These shifts of the anomalous harmonic emission times can be understood in terms of the semiclassical analytical model for the anomalous current. As we have seen, the yield minima of the $(2s)$ th harmonic is explained by the nodes of C_{2s} . When going through such a node, the real-valued amplitude C_{2s} changes sign, corresponding to a π -phase shift, which in turn will change the emission-time profiles. We confirm our interpretation by extracting the spectral phases of the anomalous harmonics from our ab-initio simulations, $\varphi^{\text{anom}}(\omega) = \arg[\mathbf{j}^{\text{anom}}(\omega) \cdot \hat{\mathbf{e}}_{\perp}]$, and present the results in Fig. 5(c). The spectral phases for H4 and H6 for the laser wavelength range $3 \mu\text{m}$ to $9 \mu\text{m}$ are in-phase with value $-\pi/2$. A sharp π -phase shift occurs for H4 at the wavelength of the yield minimum $\sim 9 \mu\text{m}$, such that H4 and H6 becomes out-of-phase from $10 \mu\text{m}$ to $16 \mu\text{m}$. This change of the spectral phase from being in-phase to out-of-phase explains the time-shift of the emissions in Figs. 5(a) and 5(b). At $\sim 16 \mu\text{m}$, the wavelength corresponding to the yield minimum of H6 in Fig. 3(c), the spectral phase of H6 shifts by π . Additional calculations confirm that the phase-shift of the anomalous HHG also occurs around the minima of a laser-intensity scan [63], again validating the robustness of our ab-initio results and our semiclassical interpretations. In addition to the emission-time shifts and the sharp spectral-phase shifts being unique characteristics of the anomalous HHG mechanism, they also suggest the potential control of anomalous harmonics by taking advantage of the sign change around the nodes of C_s .

To conclude, we have characterized the anomalous HHG mechanism in MoS_2 , by development of an ab-initio methodology that is able to decompose the different contributions to the nonperturbative microscopic current. We have identified different laser-parameter regimes wherein either anomalous or interband currents dominate, thereby resolving a recent debate on the origin of the perpendicular-polarized harmonics in terms of either the anomalous current [10–12, 14] or the interband current [20–24]. We uncovered distinct yield minima at certain field parameters, around which the subcycle emission-time profiles drastically change. Our findings open up the possibility for the experimental generation, identification and time-domain control of pure nonperturbative anomalous currents and high harmonics, which

in turn will be invaluable for high-harmonic spectroscopy of Berry curvatures [10, 11, 14], as such schemes require the generation of pure anomalous harmonics. To provide a direct example, we show in Fig. 4(a) the reconstructed $\Delta\Omega_z^k$, employing the harmonic spectrum from Fig. 2(b) in our retrieval algorithm (see SM [63]). Our work thus provides a future recipe for the experimental retrieval of Berry curvatures using HHG: (1) identify the anomalous harmonics utilizing their characteristics discussed in this work; (2) use a reliable retrieval algorithm for reconstruction.

We expect our findings to be general and directly applicable to other materials that have finite Berry curvatures. Indeed, neither the anomalous nor the perpendicular interband harmonics are prohibited by symmetry, and will generally be competing. The presence of minima in the anomalous harmonic yields are also of a general nature, since the amplitudes in Eq. (2) for varying field parameters generally take on both positive and negative values due to the Bessel functions. Finally, all our findings are facilitated by the development of an general ab-initio methodology that allows the rigorous decomposition of the currents without the construction of a globally periodic Bloch gauge, which can directly be applied to other complex systems. We expect our approach to be especially useful for ab-initio simulations of strong-field interactions in topological insulators such as Chern insulators, where a nonzero Chern number prevents the construction of a globally smooth and periodic Bloch gauge [51, 79].

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