Ultrafast spintronics with geometric effects in non-adiabatic wave-packet dynamics

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Motivated by the intriguing possibilities of steering ultrafast non-adiabatic processes through the geometric properties of bands in quantum materials by laser pulses, we extend a wave-packet transport theory, previously well-established in the adiabatic regime that intuitively captured geometric properties of bands, to the transient and non-adiabatic regime. This extension facilitates us to investigate macroscopic ways of manifesting microscopic band-geometric effects that highlight the special capability of non-adiabatic drivings not available to adiabatic drivings. These include imprinting band-geometric properties to the current rate after switching off the laser pulses and the induction of intrinsic macroscopic spin polarisation with an orientation not accessible by adiabatic processes. In particular, the microscopic geometrically-rooted intrinsic spin coherence is shown to underlie the spin-mediated parts of the macroscopic photocurrents. Through explicit calculations of an example with Rashba spin-orbit coupling, the spin-mediated part is shown to be discernible from the non-spin-mediated part in terms of the anisotropy of the photocurrents. Working principles behind the above theoretical results allegedly applicable beyond the Rashba example are distilled to inspect experimental data collected for SnSe, exhibiting considerable anisotropic effects. Consistency between theory and experiment is observed, paving the way of further exploration into the above intended direction.

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I. INTRODUCTION

Geometric and topological properties of band structures are fundamental physical assets vital for developing technologies for quantum materials. These properties can be manifested through the electronic currents, the primary observable induced in both electric transport and optoelectronic experiments. Three temporal forms of the currents are usually investigated, namely, steady-state, time-periodic, and transient. A previously established theoretical framework based on the picture of wave packets (see its introduction later) has been witnessed to be useful for intuitively understanding the band-geometric and -topological effects through steady-state as well as time-periodic currents. Particularly, with ultrashort laser pulses, the induced transient photocurrents are expected to reveal the underlying band-structure effects in a way that is not obviously visible in the steady-state and time-periodic currents. We are thus motivated here to develop extensions of this theoretically intuitive framework into the transient regime to address the band-geometric effects in laser-pulse-induced real-time photocurrents. It is well-known that nonzero Berry curve is the defining ingredient of geometric band-structure effects that have been revealed in both steady-state and time-periodic regimes \[1\]-\[11\]. To highlight the transient regime, here we will demonstrate interesting band-geometric effects by applications to systems with zero Berry curvatures.

To theoretically address the band-geometric and -topological effects on the currents, there are primarily two frameworks available in the literatures. The first is the Green-function framework \[12\]-\[14\] which had been developed for treating general properties of materials and not specifically for the above purpose. The second framework, born out of the above purpose, is the wave-packet-based transport theory \[15\]-\[17\]. In the first framework, when setting to the linear response regime, the resulting Kubo formula has been turned into TKNN formula for illustrating the role of topological effects in near-equilibrium transport and opened the venue for the studies of band topology in quantum materials \[2\]–\[18\]. Following the rigorous quantum mechanical rules, the Green-function framework had been well-established over the years and its applications related to the intrinsic band-geometric and -topological properties are mainly concentrated on tackling the extrinsic scattering effects, owing to its characteristic ability of systematically performing perturbative calculations using diagrams (see examples \[19\]-\[20\]). Despite being fully rigorous on the quantum mechanical ground, the way that band-geometric effects on transport are manifested through the heavy machinery of the Green function formulation is not as transparent as the second framework based on the picture of electron wave packets, here called the wave-packet transport theory (WPT).

The WPT is quantum mechanically rigorous on the level of a single electron as a wave packet moving among the bands. It is phenomenological on the level of electron gases as ensembles of electron wave packets. The main advantage of the WPT is its transparency of revealing band-geometric effects with clear physical intuition. It achieves this physical insight by identifying the anomalous velocity with the Berry curvatures of the bands that in turn manifest in the Hall currents \[1\]-\[15\]-\[17\]. It has found numerous applications in exploring the band-geometric effects in quantum materials \[21\]-\[27\]. Nevertheless, the application of this framework to the study of transient photocurrents induced by short laser pulses is not a straightforward and easy task. The emergence of the Berry curve in the anomalous velocity is a result of the adiabatic limit in which the applied bias to transport the electrons is relatively small in comparison to the band gaps in question. This limit enables adiabatic approximation as well as perturbation treatment of the external field. In narrowly gapped materials, featured by gaps of tens of meV at the anti-crossing \[7\]-\[28\]-\[29\], it becomes possible to break the adiabatic condition with moderate bias. Even under a constant-in-time electric field, extending the WPT to the non-adiabatic regime already met challenges due to multiple-band effects \[30\]. Still in the steady-state regime, the non-adiabatic extension of the WPT readily yields interesting results previously unseen with the adiabatic and perturbative treatments, such as non-adiabatic renormalization of valley Hall current and crystal anisotropy manifestation of non-Abelian multiple-band characters \[31\]-\[32\]. We are thus naturally motivated to further extend this non-adiabatic WPT from steady-states to the transient regime.

A natural playground of the transient and non-adiabatic regime concerning the band-geometric effects is the ultrafast spintronics with non-magnetic materials that mainly exploit spin-orbit coupling (SOC). There are a number of interesting materials where the SOC induced spin-split gaps range from tens to hundreds of meV \[33\]-\[36\] implying the possibilities of activating the non-adiabatic dynamics by attainable electric field amplitudes not more than \(100\text{meV/Å}\) (see later discussions for details of estimation). Because of the non-adiabatic nature, the aspects of the geometric properties to be focused on here can be quite different from others. The geometric properties of the bands are mathematically-defined derivatives of the band wavefunctions with respect to Bloch momentum \[17\]-\[37\]. Under small perturbing electric field, the field amplitudes can be factorised from the finite-order response coefficients in which derivatives of the band wavefunctions naturally appear \[6\]-\[38\]-\[40\]. These finite-order response coefficients, e.g., the linear conductivity as the most famous one, tell of the native properties of the materials in terms of geometric quantities as the focus of other established studies quoted above. Here in the regime of non-adiabatic drivings exceeding the validity of order-by-order analysis, the properties brought up by the drivings themselves, not just natively of the materials, become crucial for transiently steering the ultrafast electronic/spintronic processes due to the non-perturbation involvement of the laser fields. Many experiments related to SOC-supported band-geometric effects have
demonstrated exquisite capacities of manipulating ultrafast lasers, hinting feasibility of tuning the driving amplitudes to activate the non-adiabatic regime [41, 46]. Based on SOC-supported topological surface states, intriguing transient effects have been experimentally witnessed, including near unity fidelity [41], bond-specific ultrafast charge transfer [42] and polarity-tunable terahertz emission [43], just to name a few. To have a grounded starting point for exploring this relatively less charted area of non-adiabatic steering of transient dynamics using band-geometric effects in ultrafast spintronics, instead of the topological surface states with complicated geometrical properties [17], we should go back to the archetypal Rashba SOC as one foundational setup of spintronics [48–52].

For being self-content, the previously established WPT was briefly reviewed in Sec. II A. A non-adiabatic transient extension of this WPT (NADT-WPT) is introduced in Sec. II B. Details of derivations are left in the supplementary material (SM). In Sec. II C, we discuss in general possible ways that NADT-WPT can manifest band-geometric effects, especially through macroscopic observables, e.g., currents, under non-adiabatic drivings where the adiabatic WPT is not applicable. We demonstrate these effects by examples with zero Berry curvatures in Sec. III. The first example is pedagogical and realised by the Su-Schrieffer-Heeger (SSH) system in Sec. III A. The second example in Sec. III B is a prototype system with anisotropic Rashba SOC for the foundational interests of spintronics. With short laser pulses, the time domain is divided into an in-pulse (where the laser field exists) and a post-pulse region (in which the laser driving has been switched off). Our main findings are summarised here. (1): In-pulse non-adiabatic drivings allow one to manipulate post-pulse dynamics by band-geometric-enabled history-dependence effects that are not available by in-pulse adiabatic drivings. (2): The geometric effects of the SOC-governed bands can be macroscopically revealed via the intrinsic (instead of extrinsic) current-induced spin polarisation triggered by non-adiabatic drivings only. With anisotropic Rashba-SOC, such manifestation in terms of macroscopic spin polarisation is dependent on the anisotropy. (3): This anisotropic dependence furnishes a way to differentiate spin-mediated from bond-mediated processes behind the photocurrents. In Sec. IV for conclusions, we draw theoretical implications for various types of experiments. We also perform ultrafast terahertz (THz) emission experiments on SnSe showing considerable anisotropic SOC effects. Consistency between NADT-WPT results deduced independently of the Rashba SOC and experimental data is seen. Limitations and possibilities of improvements of the present theoretical development are addressed.

II. FORMULATION OF A WAVE-PACKET TRANSPORT THEORY

The WPT takes as input the band structure of the electronic material in question. The output of the theory is the electronic current \( j (t) \) under a prescribed external electric field \( E (t) \) that is in principle dependent on time \( t \). For the input, the Hamiltonian for the pristine crystalline material is prescribed as \( H (k) \) where \( k \) denotes the Bloch momentum in the Brillouin zone (BZ). By writing \( H (k) |u_n (k)⟩ = ε_n (k) |u_n (k)⟩ \) with \( n \) being the band index, the band structure is then specified by \( \{ ε_n (k) \} \) \( n \) the band energies and \( \{ |u_n (k)⟩ \} \) \( n \) the periodic parts of the Bloch wavefunctions. The band wavefunctions \( \{ |u_n (k)⟩ \} \) \( n \) collected over all the bands at any given \( k \) spans a Hilbert space called the band space. The band geometry is characterised by the properties of local variations of the band wavefunctions \( \{ |u_n (k)⟩ \} \) \( n \) over the Bloch momentum \( k \). The Berry connection \( R (k) \), which is a matrix indexed by the bands, has its elements defined by

\[
[ R_k ]_{n,m} = i \langle u_n (k) | \frac{∂}{∂k} | u_m (k)⟩ .
\]

It manifests the variations of the band wavefunctions over \( k \) and serves as a starting point to discuss band geometry.

To relate the input band structure to the output current, the WPT is constructed in two steps [17, 53]. In the first step, one considers the dynamics of a single electron wave packet carrying a charge \( −e \). Its state is characterised by its centre-of-mass (CM) coordinate in the phase space, namely, \( (x_i, k_i) \), with \( x_i \) the position and \( k_i \) the momentum, which follows

\[
\dot{x}_i = \langle [h^{-1} D_{k_i}, H (k_i)] \rangle ,
\]

\[
h \dot{k}_i = (−e) E (t) ,
\]

where \( [D_{k_i}]_{n,m} = δ_{n,m} ∂ / ∂k_i − i [R_{k_i}]_{n,m} \) is the covariant derivative (see derivations in SM-Sec. A1). In the second step, the electronic current \( j (t) \) through the electron gas as an ensemble of wave packets is constructed from collecting the velocities of all the wave packets. This is formally defined by

\[
j (t) = −e \int \mathcal{D}^k \text{Tr} (V (k) \partial_k (t)) ,
\]
where $D$ is the spatial dimension of the BZ,

$$V(k_t) = \frac{\partial \mathcal{H}(k_t)}{\partial k_t},$$  \hspace{1cm} (5)$$

is the velocity operator acting on the band space and $\varrho_k(t)$ is the one-body reduced density matrix (called the density matrix shortly afterwards) for the state of the ensemble. At time $t \leq t_0$ we assume the electron gas remains in equilibrium, namely, $\varrho_k(t \leq t_0) = \varrho^\text{eq}_k$, where

$$\varrho^\text{eq}_k = \sum_n f^{\mu,T}_F(\varepsilon_n(k)) |u_n(k)\rangle \langle u_n(k)|,$$  \hspace{1cm} (6)$$

in which $f^{\mu,T}_F(\varepsilon) = \{\exp[(\varepsilon - \mu)/k_B T] + 1\}^{-1}$ is the Fermi-Dirac distribution with $\mu$ the chemical potential, $T$ the temperature and $k_B$ the Boltzmann constant. The driving begins for $t \geq t_0$. In the integrand of Eq. (4), the relation Eq. (3) gives

$$k_t = k + \mathcal{K}_t,$$  \hspace{1cm} (7a)$$

with $k_{t=t_0} = k$ and

$$\mathcal{K}_t = -\frac{e}{\hbar} \int_{t_0}^t dt'' \mathcal{E}(t''),$$  \hspace{1cm} (7b)$$

is the net wave-vector displacement caused by the electric field accumulated from $t_0$ to $t$.

Starting from the initial state of the ensemble as Eq. (6), it is instructive to consider the intrinsic limit (no scatterings) by defining

$$\phi_{n,k}(t, t_0) := U_k(t, t_0) |u_n(k)\rangle,$$  \hspace{1cm} (8)$$

as the wave-packet state at time $t$ initiated from $|u_n(k)\rangle$ at time $t_0$, where

$$U_k(t, t') = \hat{T} \exp \left\{ -\frac{i}{\hbar} \int_{t'}^t dt'' \mathcal{H}(k_{t''}) \right\},$$  \hspace{1cm} (9)$$

is the time-ordered evolution operator with $t \geq t'$. Consequently, the ensemble of the wave packets evolves as $\varrho_k(t) \to \varrho^\text{id}_k(t)$, where

$$\varrho^\text{id}_k(t) = \sum_n f^{\mu,T}_F(\varepsilon_n(k)) |\phi_{n,k}(t, t_0)\rangle \langle \phi_{n,k}(t, t_0)|.$$  \hspace{1cm} (10)$$

The purpose of the WPT is to relate microscopic $k$-resolved observable (see, e.g., Eq. (4)) to macroscopic $k$-integrated observable (see e.g., the current Eq. (4)).

### A. Adiabatic transport of wave packets

#### 1. Formulation of adiabatic wave-packet dynamics and electronic currents

Under the adiabatic condition, we re-denote $|\phi_{n,k}(t, t_0)\rangle$ by $|\phi^{\text{ad}}_{n,k}(t)\rangle$. By driving $k_t$ sufficiently slow with small enough electric field, a wave packet starting as $|u_n(k)\rangle$ at time $t_0$ will evolve to $|u_n(k_t)\rangle$ at time $t$ with negligible occupations on $|u_{m \neq n}(k_t)\rangle$ but with the inter-band coherence between bands $n$ and $m \neq n$ accounted to the first order in the driving field. Explicitly, this so-called adiabatic driving leads to [54]

$$|\phi^{\text{ad}}_{n,k}(t)\rangle = e^{i \gamma_n(k_t)} |u_n(k_t)\rangle + \sum_{m \neq n} r_{m,n}(k_t, \mathcal{E}(t)) e^{i \gamma_m(k_t)} |u_m(k_t)\rangle,$$  \hspace{1cm} (11)$$

where

$$r_{m,n}(k_t, \mathcal{E}(t)) = \left[ \hat{R}_{k_t} \right]_{m,n} \cdot (-e) \mathcal{E}(t) \frac{(\varepsilon_m - \varepsilon_n)(k_t)}{(\varepsilon_m - \varepsilon_n)(k_t)},$$  \hspace{1cm} (12)$$
$d_n(t)$ is the dynamical phase, \( \gamma_n(k_i) = \int_{k_i}^{k_f} dk' \cdot [\mathcal{R}_{k_i}]_{m,n} \) is the geometric phase and \( [\bar{\mathcal{R}}_{k_i}]_{m,n} = e^{-i\gamma_n(k_i)} [\mathcal{R}_{k_i}]_{m,n} e^{i\gamma_n(k_i)} \). By substituting Eq. (11) to Eq. (2), the most well-known result in this first step of the WPT is

\[
\dot{x}_t = \frac{\partial \varepsilon(k_i)}{\partial k_i} - \left( -\frac{e}{\hbar} \right) E(t) \times \Omega(k_i),
\]

in which \( \varepsilon(k) = \varepsilon_n(k) \), and \( \Omega(k) \) given by

\[
\Omega(k) = \nabla_k \times [\bar{\mathcal{R}}_{k_i}]_{m,n},
\]

is known as the Berry curvature. For brevity under the adiabatic condition, the band index \( n \) is usually dropped off. Restoring the band index, we have \( \dot{x}_t \to \dot{x}_t^n = v_n^0(k_i) + v_n^1(k_i, E(t)) \). Here the first term \( v_n^0(k_i) = \partial \varepsilon_n(k_i) / \partial k_i \) due to the occupation on band \( n \) is the known as the normal velocity or group velocity. The second term, which arises from the first-order effects in inter-band coherence, given by \( v_n^1(k_i, E(t)) = (e/\hbar) E(t) \times \Omega_n(k_i) \) in Eq. (13), is known as the anomalous velocity that carries the band-geometric information as the Berry curvature. In the intrinsic limit with \( |\phi_{n,k}(t, t_0)\rangle \) in Eq. (10) replaced by \( |\phi_{n,k}^{ad}(t, t_0)\rangle \) due to the adiabatic condition, the current Eq. (14) is simply evaluated by \( j(t) = -e \int d^Dk \sum_n f_{FD}^{\mu,T}(\varepsilon_n(k)) \dot{x}^n_t \).

When one is away from the intrinsic limit, it is almost impossible to obtain the density matrix exactly without any approximations. The WPT in practice approximates \( \phi_k(t) \) by phenomenologically incorporating effects extrinsic to the pristine band structure via the scattering rates \( \{\tau_{k,1}\} \) under the relaxation-time approximation (RTA) [17, 53]. Combining the RTA for the ensemble and the adiabatic condition for single wave packet, one arrives

\[
j(t) = -e \int d^Dk \int f(k_i, t) \dot{x}_t,
\]

from Eq. (4) (see more details in SM-Sec. A3). In Eq. (15), the time-dependent distribution function \( f(k_i, t) \) is given by

\[
f(k_i, t) = f^0(k_i) + \delta f(k_i, t),
\]

in which \( f^0(k_i) = f_{FD}^{\mu,T}(\varepsilon_n(k_i)) \) accounts for the equilibrium part of the distribution and deviation from equilibrium is

\[
\delta f(k_i, t) = - \int_0^t dt' \frac{\partial f^0}{\partial \varepsilon_n(k')} P(k_i, k') \dot{x}_i^{n,\varepsilon_n^{-1}},
\]

Here,

\[
P(k_i, k') = e^{-f_{\mu}^{i,\nu}[\delta f_{\nu}^{0}]},
\]

is the probability that a wave packet carrying momentum \( k_i \) at time \( t \) will arrive at time \( t \) to the state \( k_i \) without being scattered for the whole time interval \([t, t']\) [53]. Equation (15) has been widely applied for studying band-geometric effects on transport [6, 16, 43, 49, 55, 60].

In principle, one does not need to restrict how \( E(t) \) depends on time for deploying Eq. (15) to compute electronic current \( j(t) \) as long as the adiabatic condition is satisfied. Substituting Eq. (13) for \( \dot{x}_t \) into Eq. (15) decomposes the current to \( j(t) = j^H(t) + j^L(t) \) where \( j^H(t) = -e \int d^Dk f(k_i, t) v^0(k_i, E(t)) \) is known as the Hall current coming from the anomalous velocity and \( j^L(t) = -e \int d^Dk f(k_i, t) \delta f(k_i, t) v^0(k_i) \) is the longitudinal current due to the group velocity. The most well-known result in this final step of the WPT occurs with \( D = 2 \) via the linear-response Hall conductivity \( \sigma_{xy} \) defined by \( j^H(t) = \sigma_{xy} E_y(t) \). At zero temperature, it is given by \( \sigma_{xy} = - (e^2/\hbar) \int d^2k \Omega^2(k) \), where the geometric properties of the bands rooted to the derivatives of band wavefunctions, appear in the form of the Berry curvature [2, 15, 17]. Response coefficients second orders in \( E(t) \) appear as higher derivatives in band wavefunctions, such as Berry curvature dipole, and serve as the basis for the helicity-dependent rectification of photocurrent [39] and the nonlinear Hall effect [6, 38] obtained under periodic drivings. The all-order effects from the time-period driving fields \( E(t) \) encoded in \( \delta f(k_i, t) \) have been shown to result in non-perturbation intra-band processes significant for the HHG signals in Weyl semimetals [55, 56]. However, as long as the external field \( E(t) \) is set to induce non-negligible occupations on bands not indexed by \( n \), then the transport process will not remain in the adiabatic regime, which then renders Eq. (15) inapplicable. In previous works, the above WPT (here called AD-WPT to be distinguished from NADT-WPT later) has been extended to non-adiabatic regimes but only for the steady states at \( t \to \infty \) with \( E(t \to \infty) = E \) being a time-independent constant [30, 32]. Before we continue such non-adiabatic extension of the AD-WPT to the transient regime with arbitrary time dependence of \( E(t) \), we briefly inspect the potential relevance of this regime to real materials.
2. Breaking of the adiabatic condition with moderate electric fields in the transients

The adequacy of the adiabatic condition underlying the use of AD-WPT can be assessed by the non-adiabatic measure \( r \equiv |r_{m,n}| \) given by Eq. (12) that appears in Eq. (11) describing adiabatic wave-packet dynamics. Clearly, \( r \) should be small, e.g., \( r < 2\% \), for the adiabatic condition to be held. Noting that \( r \sim k_c^{-1}(\epsilon E_{ext})/\Delta \), where \( k_c^{-1} \sim \langle |\mathbf{R}_k|_{m,n} \rangle \) and \( \Delta \) are respectively the magnitudes of the inter-band geometric connection strength and the gap, the adiabatic condition can be intentionally broken by tuning the external electric-field amplitude \( E_{ext} \) in accordance with \( k_c^{-1}/\Delta \) as an inherent material parameter. We therefore call driving scenarios \( E(t) \) that keep the smallness of \( r \) as the adiabatic drivings and otherwise non-adiabatic drivings. A definite breaking of the adiabatic condition given by \( r = 100\% \) corresponds to \( E_{ext} \rightarrow E_{na} \equiv \epsilon^{-1}\Delta/k_c^{-1} \). Note that an overwhelmingly large \( E_{ext} \) can spoil the material basis for hosting the targeted geometric properties of bands. This is concomitant to two reasons for implementing non-adiabatic dynamics particularly in the transient regime. First of all, exposing materials to strong fields for long time can inflict radiation damages. It is therefore more practical to study the non-perturbation effects of non-adiabatic dynamics particularly in the transient regime. Secondly, the sought-for intrinsic geometric effects of pure electronic origin are also anticipated to be more manifested transiently before relaxation and the dynamics of driven nuclei on longer time scales come into play. With the above consideration, to ensure the feasibility of reaching \( r = 100\% \) in real materials minding that \( E_{na} \propto \Delta \), we prescribe a limit \( \Delta \leq 200\text{meV} \) to survey the literature of first-principle calculations to obtain \( \Delta \) and \( k_c^{-1} \). While \( \Delta \) can be easily found, \( k_c^{-1} \) is usually unavailable. We then surveyed only those that provide Rashba-Dresselhaus parameters so as to estimate \( E_{na} \) using the Rashba-Dresselhaus SOC Hamiltonian to fix \( k_c^{-1} \). In Fig. 1 a number of materials with their \( E_{na} \) values are listed along the \( E_{na} \)-versus-\( k_c^{-1}/\Delta \) relation in log scale with \( E_{na} < 10\text{mV/Å} \). The diversity of the enlisted materials’ categories (see captions of Fig. 1), in addition to endorse the needs of extending the AD-WPT to NADT-WPT, also suggests us to start with simple band-structure models characterised by well-defined geometric properties to avoid complications arising from other properties, e.g., polar/ferroelectric, etc.

![Figure 1: An illustrative list of materials with \( E_{na} < 10\text{mV/Å} \), in which \( E_{na} \) (shown as the straight line against the material-inherent parameter \( k_c^{-1}/\Delta \) in log scale) is defined to be the required external field strength that reaches the value \( r = 100\% \) for the non-adiabatic measure (see the texts). The enlisted Rashba-Dresselhaus materials include polar components (blue spots: BiTeI, BiTeBr, BiTeCl [66]), ferroelectrics (red spots: GeTe [61], BiAlO3 [62]), perovskites (green spots: CH3NH3PbI3, CH(NH2)2SnI3 [63], (AMP)PbI4 [64], (FA)SnI3 [65], and 2D Janus materials (purple spots: BiTeBr, BiTeCl [66]). The ML for purple spots stands for monolayer. Here CH(NH2)2SnI3 and (FA)SnI3 are Dresselhaus-dominated while others are Rashba-dominated. The estimation is taken for their conduction-band spin splittings, whose geometric-connection effects in spin dynamics are potentially accessible by optical excitations.)](image-url)
B. Non-adiabatic transport of wave packets

Knowing the feasibility of breaking the adiabatic condition in real materials potentially of interests for band-geometric properties, we now introduce the formulation of NADT-WPT.

1. Formulation of ensemble dynamics of non-adiabatically driven wave packets

Keeping RTA without imposing the adiabatic condition on single-wave-packet dynamics results in (see details in SM-Sec. A2)

\[
\varrho_k(t) = \varrho_k(t) + \delta \varrho_k(t),
\]

(18a)

where

\[
\varrho_k(t) = \sum_i g^0_i(k_i) |u_i(k_i)\rangle \langle u_i(k_i)|,
\]

(18b)

and

\[
\delta \varrho_k(t) = -\int_{t_0}^{t'} dt' P(k_i,k_{i'}) \frac{\partial}{\partial t'} \varrho^U_k(t,t'),
\]

(18c)

in which

\[
\varrho^U_k(t,t') = U_k(t,t') \varrho_k(t') U_k(t',t),
\]

(18d)

with \( U_k(t',t) = U_k(t,t')^\dagger \) defined for \( t \geq t' \) (see \( U_k(t,t') \) defined in Eq. (9)). Here \( |u_i(k_i)\rangle \), called the \( i \)th hybridised band, is obtained by solving \( \mathcal{H}(k_i,E(t)) |u_i(k_i)\rangle = \mathcal{E}_i(k_i) |u_i(k_i)\rangle \), in which \( \mathcal{H}_{n,m}(k_i,E(t)) = \delta_{n,m}\varepsilon_n(k_i) - (1 - \delta_{n,m}) [\mathcal{R}_{k_i}]_{n,m} \cdot (-e) E(t) \) is known as the moving-frame Hamiltonian [30, 31]. The energy of the \( i \)th hybridised band is \( \mathcal{E}_i(k_i) \) and \( g^0_i(k_i) = \int_{P}^{P_T} (\mathcal{E}_i(k_i)) \). Equation (18) formally describes general non-adiabatic evolution of an ensemble of wave packets incorporating both effects due to the intrinsic band structure (through \(|u_i(k_i)\rangle\) and \( U_k(t,t') \)) and extrinsic scatterings (through \( P(k_i,k_{i'}) \) defined in Eq. (17)). By substituting Eq. (18) to Eq. (4), we find \( j(t) = \tilde{j}(t) + \delta j(t) \) with \( \tilde{j}(t) = -e \int d^Dk_i \text{Tr} [V(k_i) \varrho_k(t)] \) and \( \delta j(t) = -e \int d^Dk_i \text{Tr} [V(k_i) \delta \varrho_k(t)] \) reproducing the above mentioned Hall current and the longitudinal current respectively in the adiabatic limit (see details in SM-Secs. A2 and A3 including reproduction of Eq. (15) and the nonlinear Hall effect).

2. Differences between AD-WPT and NADT-WPT in interpretations of geometric effects

In a general WPT we picture that an electron wave packet possesses both the CM and band-pseudospin degrees of freedom (DOF). The geometric effects in wave-packet dynamics arise from the coupling between the band-pseudospin DOF and the CM DOF. The CM DOF is specified by \( (x_i,k_i) \) with \( k_i \) directly driven by the external field, Eq. (3). This coupling matrix element between the CM and the band-pseudospin DOF is explicitly given by,

\[
V_{n,m}^{\text{CM-BPSpin}}(k_i,k_i) = -\hbar \mathbf{k}_i \cdot [\mathcal{R}_{k_i}]_{n,m},
\]

(19)

for \( n \neq m \) that enables the rotation of the band-pseudospin as transitions between bands \( n \) and \( m \) via the CM motion \( \mathbf{k}_i \). This directly follows from that the band-pseudospin DOF specified by the amplitude \( \eta(t) \) is subject to \( i\hbar \eta(t) = \mathcal{H}(k_i,E(t)) \eta(t) \) (see SM-Sec A1) in which \( \mathcal{H}_{n,m}(k_i,E(t)) = V_{n,m}^{\text{CM-BPSpin}}(k_i,k_i) \) for \( n \neq m \). The consequence of interplays between the drivings (through \( k_i \) of the CM DOF) and the geometric properties of bands (through the band pseudospin) is revealed in \( \dot{x}_i \) (see Eq. (2)), the wave-packet velocity. In NADT-WPT, this CM-band-pseudospin coupling can non-perturbatively drive considerable rotations of the band-pseudospin DOF. In AD-WPT, the smallness of the non-adiabatic measure (see Eq. (12)) restricts the band-pseudospin state \( |\phi_{n,k}(t,t_0)\rangle \) to \( |\phi_{n,k}^{\text{ad}}(t)\rangle \) (see Eq. (11)). The existence of couplings among multiple bands (the fundamental reason of taking the view of band pseudospin) just modifies one band’s velocity \( \dot{x}_i^n \) from having only its own group velocity \( v_{i,n}(k_i) = \partial \varepsilon_n(k_i) / \partial k_i \) to

...
also have a Berry-curvature-governed anomalous velocity \( v'_\epsilon (k_t, E(t)) = -(−e/h) E(t) \times \Omega_n(k_t) \) as the dominant geometric effects (see Sec. [IIA]). In contrast with the availability of NADT-WPT, the multiplicity of bands, coupled by collaboration between non-adiabatic drivings and geometric connections, is not to be reduced to modification of one band’s properties. The interpretation of geometric effects relies on a more complete picture of the band-pseudospin dynamics whose particular transient non-adiabatic consequences are discussed below.

C. Geometric effects encoded in transient non-adiabatic dynamics of macroscopic observables

The main interests of studying transient currents lie in the ultrafast responses to short laser pulses. A pulsing driving field \( E(t) \) is essentially nonzero only for a finite time window \( t_0 \leq t \leq t_{\text{off}} \) and for time \( t > t_{\text{off}} \) the electric field is switched off,

\[
E(t > t_{\text{off}}) = 0.
\]  

(20)

It is therefore natural to separate the in-pulse \((t_0 \leq t \leq t_{\text{off}})\) and the post-pulse \((t > t_{\text{off}})\) time regions. Derived from Eq. [18], the geometric properties of the bands with non-perturbation effects from the transient non-adiabatic drivings can be manifested for macroscopic observables through (I): the post-pulse current rate with geometric effects embedded in the quantum acceleration shaped by transient in-pulse non-adiabatic drivings, (II): intrinsic current-induced spin polarisation and (III): spin-mediated part of the photocurrents with intrinsic spin coherence. Below we first explain these abstract points respectively, leaving concrete examples to Sec. [III]

1. Reading off geometric effects after switching off the laser pulses

The transient characters of the time-dependent current can be pronounced in its rate, namely, \( \dot{\mathbf{j}}(t) := \frac{d\mathbf{j}(t)}{dt} \). In the intrinsic limit \( \varrho_k(t) \to \varrho_k^d(t) \) (see Eq. [10]), the post-pulse current rate reads \( \dot{\mathbf{j}}(t > t_{\text{off}}) \to \dot{\mathbf{j}}^{\text{geo}}(t) \) where

\[
\dot{\mathbf{j}}^{\text{geo}}(t) = -e \int d^Dk \text{Tr}\left( \tilde{\mathcal{A}}(k_t) \varrho_k(t) \right),
\]

(21a)

with

\[
\tilde{\mathcal{A}}(k_t) := -\frac{i}{\hbar} \left[ \mathcal{V}(k_t), \mathcal{H}(k_t) \right].
\]

(21b)

See derivation details in SM-Sec. [4]. Here \( \tilde{\mathcal{A}}(k_t) \) is called the quantum acceleration since in the Heisenberg picture the commutator of an observable (here the velocity) with the Hamiltonian gives its time derivative (henceforth the acceleration). The geometric content of \( \dot{\mathbf{j}}^{\text{geo}}(t) \) (as indicated by the superscript \( \text{geo} \)) is more revealed by writing the quantum acceleration expectation value of a wave packet of with CM momentum \( k_t \) in the moving basis \( \{|u_n(k_t)\}_n \), namely,

\[
\text{Tr}\left( \tilde{\mathcal{A}}(k_t) \varrho_k(t) \right) = \frac{1}{\hbar^2} \sum_n \varepsilon_n(k_t) \sum_{m \neq n} \left( \varepsilon_m - \varepsilon_n \right) \langle k_t | \mathcal{R}_{k_t} | u_m(k_t) \rangle \langle u_m(k_t) | \varrho_k(t) | u_n(k_t) \rangle.
\]

(22)

The quantum acceleration at \( k_t \) thus is nonzero only when the band pseudospin has developed nonzero inter-band coherence \( \langle u_m(k_t) | \varrho_k(t) | u_n(k_t) \rangle \neq 0 \) for which nonzero geometric connection \( \mathcal{R}_{k_t} \) is indispensable. Since \( \dot{\mathbf{j}}(t > t_{\text{off}}) = \dot{\mathbf{j}}^{\text{geo}}(t) \), the main message conveyed by Eqs. [21] and [22] is that the current rate \( \dot{\mathbf{j}}(t) \) in the post-pulse time region \( t > t_{\text{off}} \) is a macroscopic manifestation of the microscopic geometric effect embedded in the quantum acceleration. Furthermore, such manifestion through \( \dot{\mathbf{j}}(t > t_{\text{off}}) \) is only possible for non-adiabatic drivings during \( t_0 \leq t \leq t_{\text{off}} \) but not adiabatic drivings. With extrinsic scatterings, the same conclusion about the exclusivity to non-adiabatic drivings is maintained (see SM-Sec. [4b]). A nonzero current after switching off the laser pulse implies that (i): interplays between drivings and band geometries are imprinted to the post-pulse dynamics of \( \dot{\mathbf{j}}(t > t_{\text{off}}) \) and (ii): \( \dot{\mathbf{j}}(t > t_{\text{off}}) \) carries with it the history of the past non-adiabatic drivings (see explanations in SM-Secs. [4a]).

The above (i) and (ii) have immediate implications for transient steering of electron dynamics using laser pulses in coordination with the geometric properties of the bands. It is impossible to affect post-pulse dynamics by in-pulse adiabatic drivings since the current ceases right after the pulse. In contrast, non-adiabatic drivings during the impulse time region determines post-pulse current rate through the geometric properties of the bands embedded in the
quantum acceleration as Eq. (22) (see demonstrations in Sec. III). Note that although non-adiabatic geometric effects have been studied based on Landau-Zener tunnelling scenarios [67, 68], the main focus there is the asymptotic limit, not the transient regime focused here. The essential Landau-Zener tunneling probability formula can be reached via the Markov approximation, namely, neglecting the history dependence [69]. Here we want to explore the geometric effects underlying the history dependence induced by non-adiabatic drivings.

2. Intrinsic current-induced spin polarisation (CISP)

We now turn our attention to geometric properties of bands with spin splitting induced by SOC, or briefly, SOC-geometric effects. We take the widely applied convention to single out the spin DOF with a two-band minimal description of SOC effects [36, 70, 71], namely,

\[ \mathcal{H}(\mathbf{k}) = \mathcal{T}_0(\mathbf{k}) + \mathcal{H}_{so}(\mathbf{k}), \]

where \( \mathcal{T}_0(\mathbf{k}) \) is the kinetic energy and

\[ \mathcal{H}_{so}(\mathbf{k}) = \mathbf{\Lambda}_{so}(\mathbf{k}) \cdot \sigma, \]

is the SOC in which the spin-orbit field \( \mathbf{\Lambda}_{so}(\mathbf{k}) \) is a real-valued 3D vector specified by the type of SOC under consideration and \( \sigma \) is the three-component vector of Pauli matrices. It is customary to define the spin texture, namely,

\[ \langle \sigma \rangle_n^0(\mathbf{k}) := \langle u_n(\mathbf{k}) | \sigma | u_n(\mathbf{k}) \rangle, \]

which is related to inter-spin geometric connections \( \{\mathcal{R}_n\}_{n,\bar{n}} \) where \( \bar{n} = \mp \) denotes the spin opposite of \( n = \pm \) by

\[ \frac{\partial \langle \sigma \rangle_n^0(\mathbf{k})}{\partial k_\beta} = 2 \text{Im} \left[ \langle u_n(\mathbf{k}) | \sigma | u_{\bar{n}}(\mathbf{k}) \rangle [\mathcal{R}_{n\bar{n}}]_{n,\bar{n}} \right], \]

where \( k_\beta \) is the \( \beta \)th spatial component of \( \mathbf{k} \).

Henceforth, in the absence of any external drivings the spin texture \( \langle \sigma \rangle_n^0(\mathbf{k}) \) can visualise the geometric properties of bands. In the presence of transient non-adiabatic drivings, this picture of spin texture can be extended to the time domain by defining

\[ \langle \sigma \rangle(\mathbf{k}, t) := \text{Tr} \left( \sigma \phi(\mathbf{k}, t) \right). \]

We call this non-equilibrium spin texture. For \( t \leq t_0 \), Eq. (27) describes the equilibrium spin texture, namely,

\[ \langle \sigma \rangle(\mathbf{k}, t \leq t_0) = \text{Tr} \left( \sigma \phi^0(\mathbf{k}) \right) = \sum_n f_{FD}^{\mu,T}(\varepsilon_n(\mathbf{k})) \langle \sigma \rangle_n^0(\mathbf{k}). \]

In the intrinsic limit, Eq. (10), the non-equilibrium spin texture becomes \( \langle \sigma \rangle(\mathbf{k}, t) = \sum_n f_{FD}^{\mu,T}(\varepsilon_n(\mathbf{k})) \langle \sigma \rangle_n(\mathbf{k}, t) \) where

\[ \langle \sigma \rangle_n(\mathbf{k}, t) := \langle \phi_n(\mathbf{k}, t) | \sigma | \phi_n(\mathbf{k}, t) \rangle, \]

(see definition for \( |\phi_n(\mathbf{k}, t)\rangle \) in Eq. (8)). We then see that \( \langle \sigma \rangle_n(\mathbf{k}, t) \), as an intrinsic quantity, can directly be compared to \( \langle \sigma \rangle_n^0(\mathbf{k}) \) because \( \langle \sigma \rangle_n(\mathbf{k}, t \leq t_0) = \langle \sigma \rangle_n^0(\mathbf{k}) \). So \( \langle \sigma \rangle_n(\mathbf{k}, t) \) is also a spin texture that arises in time due to the external driving. For disambiguation, \( \langle \sigma \rangle_n^0(\mathbf{k}) \) and \( \langle \sigma \rangle_n(\mathbf{k}, t) \) are called native and driven spin textures respectively. As the CM of the wave packet is driven from \( \mathbf{k} \) to \( \mathbf{k}_i \), the non-vanishing variation of the native spin texture \( \partial \langle \sigma \rangle_n^0(\mathbf{k}_i) / \partial k_i \neq 0 \Rightarrow [\mathcal{R}_{k_i}]_{n,\bar{n}} \neq 0 \) (see Eq. (26)) along the CM trajectory in BZ provides the needed CM-spin couplings to induce intrinsic spin coherence \( \langle \sigma \rangle_n(\mathbf{k}, t > t_0) \neq \langle \sigma \rangle_n^0(\mathbf{k}_i) \) as the microscopic manifestation of the SOC-geometric effects. Such effects are thus not expected for persistent spin texture [36] but could be ubiquitous for Rashba-Dresselhaus spin textures (see explanations in SM-Sec. B.1).

To macroscopically manifest the above microscopic SOC-geometric effects, the non-equilibrium spin texture \( \langle \sigma \rangle(\mathbf{k}, t) \) is directly linked to the spin polarisation \( \mathbf{S}(t) \) as a macroscopic observable, namely,

\[ \mathbf{S}(t) := \frac{\hbar}{2} \int d^Dk \langle \sigma \rangle(\mathbf{k}, t). \]

Therefore, any intrinsic geometric effects that are possibly contained in the non-equilibrium spin texture also underlies the spin polarisation by the definition of Eq. (29). For non-magnetic materials endowed with SOC, the time-reversal
symmetry (TRS) is respected. Henceforth, before the electric field driving is turned on, the spin polarisation is always zero, namely,

\[ S_{eq} = S (t \leq t_0) = 0. \]  

(30)

The possibility of applying an electric field to induce a spin polarisation via the SOC is known as the current-induced spin polarisation (CISP). Various circumstances of the CISP have been intensively studied but the focus is largely on the steady states in the adiabatic regime and not much interest from the geometric perspective is shown \[70, 72–75\]. To explore peculiarities of transient non-adiabatic aspects (as our main theme here) of the CISP, we first remind that the adiabatically induced CISP linear in the electric field does not retain properties of the intrinsic spin coherence in the resulting macroscopic spin polarisation (see details in SM-Sec. B3). This leaves the possibilities of manifesting the intrinsic spin coherence as SOC-geometric effects to non-adiabatically induced transient CISP.

3. Spin-mediated part of the photocurrents with intrinsic spin coherence.

The general separation of kinetic energy and the SOC in Eq. (23) in conjunction with Eq. (5) leads to a natural decomposition of the velocity matrix as

\[ V (k_t) = V^K (k_t) + V^{so} (k_t), \]  

(31a)

where

\[ V^K (k_t) := \frac{\partial T_0 (k_t)}{\partial \hbar k_t}, \]  

(31b)

comes from the kinetic energy and

\[ V^{so} (k_t) := \frac{\partial H_{so} (k_t)}{\partial \hbar k_t}. \]  

(31c)

is due to the SOC. Substituting Eq. (31) to Eq. (4) further decomposes the electronic current to

\[ j (t) = j_K (t) + j_{so} (t), \]  

(32a)

where

\[ j_K (t) = (-e) \int d^D k T \{ V^K (k_t) \varrho (k_t) \}, \]  

(32b)

and \[ j_{so} (t) = (-e) \int d^D k T \{ V^{so} (k_t) \varrho (k_t) \} \] which by Eq. (24) can be alternatively written as

\[ j_{so} (t) = (-e) \sum_{\beta \in \{x,y,z\}} \int d^D k \frac{\partial \Lambda_{so,\beta} (k_t)}{\partial \hbar k_t} \langle \sigma_\beta \rangle (k, t), \]  

(32c)

where \( \langle \sigma_\beta \rangle (k, t) \) and \( \Lambda_{so,\beta} (k_t) \) are the \( \beta \)th components of \( \langle \sigma \rangle (k, t) \) (defined in Eq. (27)) and \( \Lambda_{so} (k_t) \) respectively. Since \( j_{so} (t) \) the spin-mediated part of the photocurrent for obvious reasons. The other part of the current, \( j_K (t) \), that exists without any consideration of spin is due to bonding among neighbouring unit cells. Here we call it the bond-mediated part. Since the intrinsic part of the non-equilibrium spin texture \( \langle \sigma \rangle (k, t) \) contains the SOC-geometric effects, the spin-mediated current \( j_{so} (t) \) as a macroscopic observable can potentially manifest such effects.

III. EXAMPLES

Here in this section, we illustrate the above abstract points (I-III) of Sec. IIC with concrete examples of band structures. The examples addressed here are all with zero Berry curvatures so as to focus on the manifestation of geometric properties through the transient non-adiabatic dynamics.
A. Transient non-adiabatic manifestation of geometric effects in SSH system

Hereby with SSH system we demonstrate the point (I) outlined in Sec. IIC1. The SSH system consists of bonded sublattices arranged periodically in one dimension with $D = 1$ so $k \rightarrow k$, $\mathcal{K}_1 \rightarrow \mathcal{K}_2$, and $\dot{j} (t) \rightarrow \dot{j} (t)$. By the simplicity of 1D BZ, the Berry curvature is zero \[26\]. The SSH Hamiltonian $\mathcal{H}_{SSH} (k)$ can be seen as a case of Eq. \[23\] specified by $\mathcal{T}_0 (k) = 0$ and $\mathcal{A}_\sigma (k)$ replaced by $\mathcal{A}_{SSH} (k) = (b_1 + b_2 \cos (ka), b_2 \sin (ka), 0)$ in which $b_1$ and $b_2$ are bonding strengths and $a$ is the lattice constant \[77\]. The relations among spin-geometric effects, intrinsic spin coherence, native and driven spin textures, enunciated in Sec. IIC2 supplemented by SM-Secs. B1 and B2 equally apply here via the substitution of spin by sublattice pseudospin.

Here the native sublattice-pseudospin texture determined by $\mathcal{A}_{SSH} (k)$ dictates that $\langle \sigma \rangle_0 (k) \cdot \hat{z} = 0$. The quantum acceleration defined by Eq. (21b) gives $\dot{\mathcal{A}} (k_t) = -2a\hbar^{-2} (b_2^* + b_1 b_2 \cos k_t a) \sigma_z$. So Eq. (21) and $\dot{\mathcal{A}} (k_t) \propto \sigma_z$ for the SSH system together signify that the non-vanishing of the post-pulse macroscopic current rate $\dot{j} (t > t_{off}) = \dot{j}_{geo} (t)$ testifies microscopic intrinsic pseudospin coherences $\langle \sigma \rangle_n (k, t) \neq \langle \sigma \rangle_0 (k_t) \cdot \hat{z} = 0$. We demonstrate these effects explicitly in Fig. 2 with laser pulses $E (t)$ subject to Eq. (20) \[78\] for pulse setting and units. As expected by the sub-point (i) of (I), different geometric properties (here set up by choices of $b_1$ and $b_2$) of the bands can leave their imprints to the post-pulse current rates. This is exemplified in Fig. 2(a) where the two curves of $\dot{j} (t > t_{off})$ differ in all three attributes of an oscillating current rates, namely, amplitudes, frequencies and phases. By setting the two compared band structures to share the same gaps but different geometric properties, encoded by $b_1 < b_2$ and $b_1 > b_2$ also as different topologies, in Fig. 2(b), the two curves of $\dot{j} (t > t_{off})$ appear to have the same frequency but considerably different amplitudes. To illustrate the sub-point (ii) of (I), driving-history dependence, two pulses of the same duration but different central frequencies are depicted in Fig. 2(c) and the two corresponding current rates are shown in Fig. 2(d). The in-pulse transient oscillations displayed by the two curves of $\dot{j} (t < t_{off})$ driven by these two pulses obviously differ in oscillation frequencies. Nevertheless, for the post-pulse oscillations $t > t_{off}$, they differ more apparently in the oscillation phases/amplitudes but with equal periods, attesting driving-history dependence as geometric effects explained formally in more details in SM-Sec. B2.

Figure 2: Imprints of in-pulse interplays between drivings and band-geometry on post-pulse current rates ((a), (b)) and driving-history dependencies ((c), (d)). The vertical dotted line in each plot marks the time the pulse ceases $t_{off}$. The blue dotted line in (a), whose value goes by the right vertical axis, is the waveform $K_2$ of the pulse (see more details in \[78\]) with the amplitude $\mathcal{K}^* = 0.05 \pi /a$ and central frequency $\omega = \omega^*$. (a): Different settings of bonds $(b_1, b_2)$ (see legend) under the same topology $b_1 < b_2$ show distinct post-pulse oscillation phases and amplitudes. (b): Distinct topologies (see legends for both $b_1 < b_2$ and $b_1 > b_2$) with the same dispersion show the same post-pulse oscillation periods but with sizably different amplitudes. In (c), two different pulses differing only in central frequencies (see legend) otherwise identical (with $\mathcal{K}^* = 0.2 \pi /a$) are portrayed. Correspondingly in (d), two different current rates (with the same band structure specified by $(b_1, b_2)$ in the legend) are plotted. During in-pulse time region, different driving frequencies naturally result in different transient oscillation frequencies of the current rates. No difference in oscillation frequencies show up after the pulse is off (see marks of the equal periods). However, the difference in driving histories shows up as different phases and amplitudes of post-pulse oscillations.
B. SOC-geometric effects in ultrafast opto-spintronics with Rashba SOC

Hereby we demonstrate the points (II) and (III) abstracted in Secs. IIC2 and IIC3 with Rashba SOC. The Rashba spin-orbit field is defined in \( D = 2 \). This endows the system with the possibility of in-plane anisotropy \([33, 34, 79, 80]\) whose interesting physical effects have been witnessed, such as the fermi surface fragmentation \([70]\) and modulation of optical conductivities under continuous irradiation \([71]\). We therefore include the anisotropy in our following investigations.

As has been established in Refs. \([71, 80]\), the Rashba Hamiltonian with effective-mass anisotropy has its kinetic energy part given by

\[
T_0 (k) = \frac{\hbar^2 k_x^2}{2m_x} + \frac{\hbar^2 k_y^2}{2m_y},
\]

in which \( m_x/y \) is the effective mass along the \( x/y \)-direction and its SOC part specified by the spin-orbit field given by

\[
\Lambda_{so} (k) = \alpha_R [\lambda \times \hat{z}],
\]

in which \( \alpha_R \) is the Rashba coefficient and \( \lambda = \left( \begin{array}{cc} x & 0 \\ 0 & y \end{array} \right) \) anisotropically rescales the components \( k_x \) and \( k_y \) of the Bloch momentum \( k = k_x \hat{x} + k_y \hat{y} \) to \( \lambda_K = r_X k_x \hat{x} + r_Y k_y \hat{y} \) with \( r_X = \sqrt{m_y/m_x} \) and \( r_Y = \sqrt{m_x/m_y} \). To simplify the description of the anisotropy, we define

\[
r_A := m_y/m_x.
\]

By setting \( r_A = 1 \), Eq. (34) reduces to the familiar isotropic Rashba SOC with \( \Lambda_{so} (k) = \alpha_R \hat{k} \times \hat{z} \). When the anisotropy is present, namely, \( r_A \neq 1 \), the system then possesses \( C_{2v} \) symmetry \([70]\) in which the \( x-z \) and \( y-z \) planes are the two mirror planes. For brevity in the present \( D = 2 \) analysis, we call the \( x- \) and \( y- \) axes the mirror lines. For investigating the effects of the anisotropy, we fix the orientation of \( E (t) \) to be independent of time, namely, \( E (t) = \hat{e} E (t) \) with \( \hat{e} = \hat{x} \cos \varphi_e + \hat{y} \sin \varphi_e \), specified by an angle \( 0 \leq \varphi_e \leq \pi \). The anisotropy can then be revealed through the dependence on \( \varphi_e \). Arbitrary vectorial physical quantity, e.g., the macroscopic spin polarisation \( \hat{S} \), can then be decomposed as \( \hat{S} = \hat{e} S^\parallel + (\hat{z} \times \hat{e}) S^\perp + \hat{z} S^z \) where the symbols \( \perp \) and \( \parallel \) stand for in-plane transverse and longitudinal components to track anisotropic effects. Regardless whether or not \( r_A = 1 \), the Rashba spin-orbit field \([34]\) straightforwardly gives zero Berry curvature according to Eq. (14). So the SOC-geometric effects here are not caused by the Berry curvature.

1. SOC-geometric effects behind transient non-adiabatic CISP

The Rashba SOC given by Eq. (34) shows the native spin texture \( \langle \sigma \rangle^0_{\pm} (k) = \pm \lambda \hat{k} \times \hat{z} / |\lambda K| \) with \( \langle \sigma \rangle^0_{\pm} (k) = 0 \). A nonzero out-of-plane driven spin texture \( \langle \sigma \rangle^\pm_\pm (k, t) \neq 0 \) therefore indicates the existence of intrinsic spin coherence raised by the external field. This also raises the possibility of getting \( S^z (t) \neq 0 \) whose existence then represents a macroscopic manifestation of the microscopic intrinsic spin coherence rooted to the Rashba-SOC geometric effects. Through a detailed investigation using Eq. (18) including both the intrinsic limit and with extrinsic scatterings (see SM-Secs. C1 and B3), we found that although both adiabatic and non-adiabatic drivings can induce microscopic spin coherence \( \langle \sigma \rangle^\pm_\pm (k, t) \neq 0 \), a non-vanishing macroscopic out-of-plane CISP is a result of non-adiabatic drivings that break the anisotropy-supported mirror symmetry \( \varphi_e \neq 0, \pi/2 \) (see a demonstration in Fig. 3a)). We summarise these in the table:

| \( \langle \sigma \rangle^\pm_\pm (k, t) \neq 0 \) | adiabatic | non-adiabatic | \( S^z (t) \neq 0 \) | adiabatic | non-adiabatic |
|---|---|---|---|---|---|
| \( r_A = 1 \) | yes | yes | \( r_A = 1 \) | no | no |
| \( r_A \neq 1 \) | yes | yes | \( r_A \neq 1 \) | no | yes |

Indeed, the association of anisotropic characteristics, i.e., dependence on the orientation angle \( \varphi_e \), with the geometric properties of bands has already been hinted by the coupling between the wave-packet CM and its band pseudospin in Eq. (19). There \( \hbar k \| \hat{e} \) participates in this scalar product by its orientation relative to the geometric connection. This hint has been brought to light explicitly by the non-adiabatic transient out-of-plane CISP effects realised with anisotropic Rashba SOC.

2. Anisotropic characteristics differentiated between spin-mediated and bond-mediated parts of the photocurrent

The manifestation of SOC-geometric effects in the out-of-plane spin polarisation through non-adiabatically breaking the mirror symmetry \( \langle \hat{e} \| \hat{x}/\hat{y} \rangle \) has further implications for the spin-mediated processes in the photocurrents as
discussed in Sec. IIC3. We make the dependence on \( \hat{e} \) (specified by \( \varphi_e \)) explicit by writing \( j(t)|_{\varphi_e} = j(t, \varphi_e) \). It becomes convenient to define the anisotropic asymmetry, namely,

\[
\Delta \delta \varphi j_{K/so}^\alpha (t) := j_{K/so}^\alpha (t, \delta \varphi) - j_{K/so}^\alpha (t, \pi/2 - \delta \varphi) .
\] (36)

It quantifies the efficacy of the anisotropy in the \( \alpha \)-component of the bond/spin-mediated current (see Eq. (32)) under the circumstance of breaking the mirror symmetry by deviating of \( \hat{e} \) from both of the mirror lines by an angle \( \delta \varphi \). Analysing bond/spin-mediated parts of the photocurrent respectively with Eq. (36) (see details in SM-Sec. C2) shows that

\[
|\Delta \delta \varphi j_{\perp}^\perp (t) | \ll |\Delta \delta \varphi j_{\parallel}^\parallel (t) | \approx |\Delta \delta \varphi j_{\perp}^\parallel (t) | .
\] (37a)

\[
|\Delta \delta \varphi j_{\perp}^\parallel (t) | \approx |\Delta \delta \varphi j_{\perp}^\parallel (t) | \gg |\Delta \delta \varphi j_{\parallel}^\parallel (t) | .
\] (37b)

We make two remarks on Eq. (37b). First, for small \( \delta \varphi \), \( \Delta \delta \varphi j_{\perp}^\parallel (t) \) is determined by \( S^\parallel \) (see Eq. (C15b)). Second, under non-adiabatic drivings, \( S^\parallel (t, \varphi_e) \) rises simultaneously with \( S^\perp (t, \varphi_e) \) (see analytical indications in SM-Sec. C1 and numerical demonstrations in Fig. 3(b) that accompanies Fig. 3(a)). We already know \( S^\perp (t, \varphi_e) \) signifies SOC-geometric effect from previous discussions. Henceforth, the above two remarks plus the dominance of \( \Delta \delta \varphi j_{\perp}^\parallel (t) \) in \( \Delta \delta \varphi j_{\perp}^\parallel (t) \) indicated by Eq. (37b) is also a signature of SOC-geometric effects. The result Eq. (37) tells that spin-mediated part \( j_{\perp} \) (t) can be differentiated from the bond-mediated part \( j_{K} \) (t) of the photocurrent \( j (t) = j_{K} (t) + j_{so} (t) \) by their respective dominances in the anisotropic asymmetry of the \( \perp \) and \( \parallel \) components, see Fig. 3(c) (for Eq. (37b)) and (d) (for Eq. (37a)). This requires to discern \( \perp \) from \( \parallel \) components of the full photocurrent in terms of \( \varphi_e \)-dependence characters. Such discernment can be arrived by exploiting the anisotropy-furnished mirror symmetry, namely,

\[
j_{\perp} (t, M\hat{e}) \neq j_{\perp} (t, \hat{e}) , j_{\parallel} (t, M\hat{e}) = j_{\parallel} (t, \hat{e}) ,
\] (38)

in which \( M\hat{e} \) reflects \( \hat{e} \) by either of the two orthogonal mirror lines (see details in SM-Sec. C2).

IV. CONCLUSIONS

A. Summary

With the development of a NADT-WPT as an extension of well-established AD-WPT, we have explored three distinct ways of manifesting band-geometric effects in macroscopic observables for transient non-adiabatic drivings, namely, the current rates, the spin polarisation, and the spin-mediated part of the photocurrents. We have shown geometric properties of bands leave their imprints on the patterns of current rate dynamics after turning off the laser pulses. This is through the history-dependence effects enabled by non-adiabatic in-pulse drivings via the mechanism of quantum acceleration. We have demonstrated the above mechanisms using the example of the SSH system and anticipate similar phenomena to be found in other systems as well [81]. The macroscopic spin polarisation is revealed to contain band-geometric effects that are exclusive to non-adiabatic drivings due to its embedding of intrinsic spin coherences. This contrasts with the adiabatic CISP that is proportional to the relaxation time characterising extrinsic scatterings. With Rashba SOC defined with in-plane native spin texture, we demonstrated how the out-of-plane CISP signifies such SOC-band-geometric effects albeit restricted by the SOC anisotropy. The spin-mediated part of the photocurrent signifies SOC-band-geometric effects due to the same reason of its association with the intrinsic spin coherence. The dependence of CISP as well as of the photocurrents on the anisotropy of the SOC furnishes a way to differentiate spin-mediated from the bond-mediated parts in the full photocurrent by the former’s distinct dominance in the anisotropic asymmetry of the transverse component of the photocurrent. The above spin-related effects, rooted to intrinsic spin coherence, have also been examined including extrinsic scatterings via NADT-WPT. The salient features of their manifestations are not altered by the extrinsic scatterings, owing to the transient dominance of intrinsic geometric properties induced by non-adiabatic drivings.

B. Implications for experiments

With the application of ultrafast laser pulses to quantum materials, three types of detections are commonly seen in experiments, namely, the time- and angle-resolved photoemission spectroscopy (Tr-ARPES), time-integrated elec-
tronic currents at contacts, and time-resolved THz emissions. We have investigated both microscopic momentum-resolved electron dynamics and macroscopic observables like the current. The former has been experimentally targeted in general through the Tr-ARPES down to subcycle time resolutions. Retrieving band structure information from the electronic currents excited by laser pulses has also been experimentally practiced. For time-resolved THz emissions, within the framework of electromagnetism, the emitted electric fields $E_{em}(t)$ have been shown to be proportional to $dj(t)/dt$. This provides a link between experimental signals and the present theory which computes $j(t)$ and $dj(t)/dt$. So broadly speaking, the present theoretical endeavour is related to all three types of experiments. More specifically, concerning the particular geometric effects and their manifestations through macroscopic observables, there are subtleties and caveats for each type of the experiments.

Nevertheless, despite the complexities arising from the band compositions for band-geometric effects, the present results comprehending Secs. [III C 2] [III C 3] and [III B] readily furnish implications relevant for experimental studies of certain aspects of SOC effects (discussed below) within the broader context of spintrons. Notably, the decomposition $j(t) = j_{so}(t) + j_{K}(t)$, Eq. [32], is held not only for two-band descriptions. It is inherited to any Hamiltonian describing an electron moving in periodic potentials with an SOC part and the other part that exists readily without SOC. Recall from Sec. [III C 3] that the spin-mediated part $j_{so}(t)$ differentiates itself from the bond-mediated part $j_{K}(t)$ of the photocurrent $j(t)$ readily in terms of their microscopic underpinning processes, without referring to specific type of SOC. With the Rashba SOC in Sec. [III B], we showed that $j_{so}(t, \varphi)$ and $j_{K}(t, \varphi)$ are distinctively dominated by $j_{so}^\perp(t, \varphi)$ and $j_{K}^\parallel(t, \varphi)$ in terms of the anisotropic asymmetry Eq. [37]. Apart from the result of Eq. [37], that is specific to the Rashba SOC example of Sec. [III B], the above NADT-WPT results generally testify three
points that are independent of the SOC type. (i): The spin and the usual electronic bond mediate fundamentally different processes behind the photocurrents. (ii) The responses along $\parallel$ and $\bot$ directions are induced by quite different mechanisms (differentiated by Eq. (38) as an example) so that (iii): their respective ways of encoding spin-mediated and bond-mediated processes, e.g., through anisotropic characteristics, are also different, exemplified by Eq. (37). These theoretical implications indicate the latent potency of structural anisotropy in experimentally revealing the underlying spin-related processes. Hereby we perform THz-emission experiments on SnSe whose anisotropic effects have already received much attention in thermoelectric properties [83] but not yet fully explored for the spintronic processes with implications from NADT-WPT.

The 3D bulk single-crystal SnSe has an orthorhombic structure which naturally possesses a termination plane (called the sample plane) with two orthogonal non-equivalent crystal axes, similar to the two orthogonal mirror lines addressed in Sec. III B. To relate to the above points (i-iii), we collected THz emissions with both $\parallel$ and $\bot$ components generated by linearly polarised (LP) pulses with various polarisation angles (to map out its anisotropic characters) together with those induced by circularly polarised (CP) pulses. For details of experiments, see SM-Secs. D2a. Deductions made purely from the CP data and that purely from the LP data aided by NADT-WPT lead to the same conclusion that the processes on the sample plane dominate the detected signals (see SM-Sec. D2b aided by SM-Sec. D2d). An analysis of these data without a priori assumption of the SOC type shows consistency between the experimental data and the points (i-iii) above (details in SM-Sec. D2c). This consistency result on one hand supports the meaningful existences of the spin-mediated and the bond-mediated parts of the photocurrents. On the other hand, it also helps to illustrate the potential feasibility of exploiting the anisotropy for spintronic interests through the additional the use of LP pulses, complementary to the familiar association of SOC effects to CP-differentiated responses in THz-emission experiments.

C. Limitations and possibilities of improvements

This work shows one step toward the big question of how band-geometric properties play their roles in transient and non-adiabatic steering of micro/macrosopic processes. We proposed a wave-packet-based framework to analyse the driving-dominated revelation of band-geometric effects. At this stage, we then have only explicitly calculated results from two-band systems. More refined band-structure and relaxation times customised for specific materials can always be incorporated as inputs to NADT-WPT for future studies of these materials. We also have only compared two big categories of drivings, namely, adiabatic and non-adiabatic drivings. This is because how band-geometric properties are carried with adiabatic transport is readily well known. The logical first step is then to identify manifestations of geometric effects that are exclusively available for non-adiabatic drivings. As experimental techniques of tailoring ultrafast laser pulses are readily advanced, comparison among non-adiabatic transient drivings of different characters await.

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Supplementary Materials for Ultrafast spintronics with geometric effects in non-adiabatic wave-packet dynamics

For being self-content, in Sec. A we give a derivation of how the previously established WPT (or AD-WPT) is extended to the transient and non-adiabatic regime (NADT-WPT) as the new development here, supplementing primarily the main text contents of Sec. II and III A. In Sec. III B of this SM, we link the well-established concepts of spin texture to wave-packet dynamics for enunciation of spin-related geometric properties of bands. These links underlie the primary discussions abstracted in Sec. III C and demonstrated with concrete examples in Sec. III D of the main text. We have briefly in Sec. III D of the main text the primary results of the particular example of anisotropic Rashba SOC. Here in Sec. IV of this SM, we supplement these results by systematic investigations using the NADT-WPT applied to the Rashba systems under consideration. We describe implications to experiments with ultrafast lasers of several types in general in Sec. IV A. More specifically we detail the THz-emission experiments on SiSe with the subsequent analysis of the data in Sec. IV B. The SM-Sec. IV underlies the discussions in Sec. IV of the main text.

Appendix A: A wave-packet transport theory

As mentioned in the main text, the WPT is constructed in two steps [17, 53]. In the first step, a single electron wave packet is considered (detailed in Sec. A1) while for the second step, macroscopic current in an electron gas is derived from an ensemble of wave packets (detailed in Sec. A2). While reproduction of steady-state AD-WPT from the non-adiabatic but static extension of AD-WPT has been detailed in [30], here the reproduction of time-dependent AD-WPT results (including the well-known nonlinear Hall effect [6]) from NADT-WPT is described in Sec. A3. In Sec. A4 the real-time non-adiabatic current formula from NADT-WPT is analysed for the post-pulse time region.

1. Dynamics of a single wave packet with multiple bands

The equations of motion followed by a single wave packet with multiple bands, namely, Eqs. (2) and (3) of the main text, have been derived using variational-principle on wave-packet Lagrangian in [31, 55]. They have been re-derived without using the Lagrangian-based variational formalism in Ref. [30]. In this subsection, we give a short summary of the much more detailed derivation given in [30] to obtain Eqs. (2) and (3) and take the adiabatic limit to arrive Eq. (13) given in the main text.

The starting point is an electron with charge $-e$ moving in a periodic potential described by the Hamiltonian $H(r, p, \sigma)$ where $r$, $p$ and $\sigma$ are the coordinate, momentum and Pauli operators for the spin respectively. Under a spatially uniform electric field $E(t) = -A(t)$ generated by a vector potential $A(t)$ with the minimum coupling principle, $p \rightarrow p - (-e)A(t)$, one obtains a time-dependent Schrödinger equation $H(t)|\Phi(t)\rangle = i\hbar|\dot{\Phi}(t)\rangle$ with the time-dependent Hamiltonian $H(t) := H(r, p - (-e)A(t), \sigma)$. Under the Born–von Karman boundary condition, we can take the ansatz: $|x, s|\Phi(t)\rangle = \sqrt{1/\mathcal{N}} e^{ik_s x} |x, s|\chi_t\rangle$ in which $r|x, s\rangle = x|s\rangle$, $\sigma \cdot \hat{n}|x, s\rangle = \pm |x, \pm\rangle$ for an arbitrarily prescribed real 3D unit vector $\hat{n}$. Here $|\chi_t\rangle$, normalised to unity within a primitive cell, is a cell-periodic function, namely, $|x, s|\chi_t\rangle = |x + R, s|\chi_t\rangle$ with $R$ being an arbitrary lattice vector and $1/\sqrt{\mathcal{N}}$ is a normalisation constant. In the space of periodic functions, we then have $i\hbar|\dot{\chi}_t\rangle = \mathcal{H}(k_t)|\chi_t\rangle$ where $\mathcal{H}(k_t) = H(r, p + ik_t, \sigma)$ in which $ik_t = h\nabla - (-e)A(t)$. The time derivative $i\hbar\dot{k}_t = (-e)\left(-\dot{A}(t)\right)$ immediately becomes Eq. (13) of the main text. We now derive Eq. (2) of the main text. The velocity operator is defined by $v(t) := (-1/\hbar)[r, H(t)]$ so the wave packet velocity is given by $\dot{x}_t = \langle \Phi(t)|v(t)|\Phi(t)\rangle$. By the use of $\langle x,s|\Phi(t)\rangle = \sqrt{1/\mathcal{N}} e^{ik_s x} |x, s|\chi_t\rangle$ and the periodicity, it reads $\dot{x}_t = \langle \chi_t|V(k_t)|\chi_t\rangle$ in which $V(k_t)$ is defined by Eq. (5) of the main text. To pronounce the geometric aspects, we expand $|\chi_t\rangle = \sum_{n,m} \eta_n(t) e^{i\gamma_n(k_t)} |u_n(k_t)\rangle$ which then leads to $\dot{x}_t = \sum_{n,m} \eta_n(t) e^{-i\gamma_n(k_t)} \left[h^{-1}\mathcal{D}_{k_t}, \mathcal{H}(k_t)\right]|u_n(k_t)\rangle$ as Eq. (2) of the main text.

The more familiar expression for $\dot{x}_t$ in which the Berry curvature appears along with the anomalous velocity [17] (as Eq. (13) in the main text) can be derived straightforwardly from the above by first segregating the bands $\{|u_n(k_t)\rangle\}_{n}$ into two sets, namely, the active and the remote manifolds, denoted by $a$ and $r$ respectively. The definition is that during the driving processes, the occupations on the remote manifold $\{u_n(k_t)\}_{n\in r}$ are negligible while those on $\{u_n(k_t)\}_{n\in a}$ are not. With the time-dependent basis of $\{|u_n(k_t)\rangle\}_{n}$, the Schrödinger equation $i\hbar|\dot{x}_t\rangle = \mathcal{H}(k_t)|\chi_t\rangle$ is turned into $i\hbar\dot{q}_t = \mathcal{H}(k_t, E(t))q(t)$ in which the $n$th component of the complex vector $q(t)$ is $q_n(t)$ and the moving-frame Hamiltonian $\mathcal{H}(k_t, E(t))$ described below Eq. (18) of the main text naturally appears. By taking the
inter-manifold matrix elements $\mathcal{H}_{n,m}(k, E(t)) = -[\mathcal{R}_{k}]_{n,m} \cdot (-e) E(t)$ in which $n \in a$ and $m \in r$ or $n \in r$ and $m \in a$ as perturbation to its first order, Eq. (2) of the main text becomes

$$\dot{x}_{\alpha t} = \langle [\hbar^{-1} D_{k\alpha t}, \mathcal{H}_{a}(k)] \rangle_{a} - \sum_{\beta} \langle \mathcal{F}^{\alpha \beta}(k) \rangle_{a} k_{\beta t},$$  \hspace{1cm} (A1)$$
in which the active-manifold average $\langle \mathcal{M}(k) \rangle_{a} := \sum_{n, m \in a} \eta_{n}^{a}(t) e^{-i\gamma_{n}(k)} [\mathcal{M}(k)]_{nm} \eta_{m}(t) e^{i\gamma_{m}(k)}$ has been defined for arbitrary matrices $\mathcal{M}(k)$. Here in Eq. (A1), $\dot{x}_{\alpha t}$ and $k_{\beta t}$ are respectively the $\alpha$th and the $\beta$th spatial components of $\dot{x}_{t}$ and $k_{t}$ and the definition

$$\mathcal{F}^{\alpha \beta}(k) := \frac{\partial [\mathcal{R}_{k\beta t}]}{\partial k_{\alpha t}} - \frac{\partial [\mathcal{R}_{k\alpha t}]}{\partial k_{\beta t}} - i \mathcal{R}_{k\alpha t}, \mathcal{R}_{k\beta t},$$  \hspace{1cm} (A2)$$
known as the non-Abelian Berry-curvature has been applied. The simpler results Eqs. (13) and (14) in the main text are obtained from Eqs. (A1) and (A2) by assuming there is only one band indexed by $n$ in the active manifold. Hereby we complete the derivation for the equations of motion followed by a single wave packet present in the main text.

2. Dynamics of ensemble of wave packets

We now move to the second step of the WPT in which we consider an ensemble of wave packets. We derive Eq. (18) of the main text by extending the kinetic theory readily framed in Refs. [17, 53] to account for scattering effects. For being pedagogical, we discuss consecutively three cases (a-c). (a): The band gaps and the external electric fields are set to satisfy the adiabatic condition. (b): The band gaps and the external electric fields are not set to satisfy the adiabatic condition but the external electric fields are assumed to be static at times of interests for steady states. (c): The external electric fields are time-dependent and we are interested in the transient regime. We start by describing the formulation in the kinetic theory for electron gases, that is common to all the three cases. We then briefly review the specification to situations (a) in Sec. A2a (well established in Refs. [17, 53]) and (b) in Sec. A2b (detailed in Ref. [33]). After this, how the specification to (c) can be done then becomes obvious in Sec. A2c. Since the main focus of the present research is on the time-dependent phenomena, in Sec. A3 we show that under periodic driving with small amplitudes such that the adiabatic condition is valid, the second-order response calculated using the density matrix given by Eq. (18) reproduces the well-established quantum nonlinear Hall effect of Ref. [9].

The state of the ensemble is described by the density matrix $\rho_{k}(t)$ collected over all $k$’s. Recall that previous discussions for the dynamics of a single wave packet do not include any scattering effects. So a single wave packet with CM momentum $k = k_{t_0}$ initially in an arbitrary pseudospin state $|\chi = \chi_{t_0}\rangle$ (collectively denoted as $(k, \chi)$) will evolve to the state $(k_{t}, \chi_{t})$ with $|\chi_{t}\rangle := U_{k}(t, t_{0}) |\chi_{t_{0}}\rangle$ at any later time $t > t_{0}$. The matrix element $\langle \chi_{t_{0}} | \rho_{k}(t) | \chi_{t_{0}} \rangle$ gives the ensemble-averaged occupation on $|\chi_{t_{0}}\rangle$ from the initial ensemble $\rho_{k}(t_{0})$. Imposing scattering-free dynamics to all wave packets in the ensemble results in $\langle \chi_{t} | \rho_{k}(t) | \chi_{t} \rangle = \langle \chi_{t_{0}} | \rho_{k}(t_{0}) | \chi_{t_{0}} \rangle$. We now consider how scattering events affect the occupation $\langle \chi_{t} | \rho_{k}(t) | \chi_{t} \rangle$. Due to scattering, only a part of this occupation is actually contributed by a wave packet that has been coherently driven to the state $(k_{t}, \chi_{t})$. The effects of scatterings are taken into account by the relaxation-time approximation (RTA) which consists of two assumptions (i) and (ii) that we will individually introduce in appropriate places. The assumption (i) is that the scattering rate is independent of the form of the electron distribution function. Using (i), we then denote by $1/\tau_{k}$ as the scattering rate for a wave packet carrying momentum $k$. We denote by $P(k_{t}, k_{t'})$ the probability that a wave packet in the state $(k_{t}, \chi_{t})$ at time $t$ can evolve to the state $(k_{t'}, \chi_{t'})$ at time $t + t'$ without being scattered away. By a general argument, $P(k_{t}, k_{t'})$ is found to be given by Eq. (17) of the main text [53, 86]. Henceforth, the contribution to $\langle \chi_{t} | \rho_{k}(t) | \chi_{t} \rangle$ by the above field-driven process is given by

$$g_{\chi_{t}}^{\text{drift}}(k, t) = \int_{t' = t_{0}}^{t'} dt' \; g_{\chi_{t'}}(k_{t''), t') \; P(k_{t}, k_{t''}),$$  \hspace{1cm} (A3)$$
where $d_{\chi_{t}}(k_{t''}, t'')$ is the amount occupation gained into the state $(k_{t''}, \chi_{t'})$ in an infinitesimal time interval of length $dt'$ around time $t'$. Note that at $t = t_{0}$, $g_{\chi_{t}}^{\text{drift}}(k, t_{0}) = 0$ by the definition of Eq. (A3). So if the initial equilibrium readily has a nonzero occupation on $(k, \chi)$ (so it can be coherently driven to occupy $(k_{t}, \chi_{t})$), this source of contribution to the occupation is not accounted for by Eq. (A3). By including also this contribution, we obtain

$$\langle \chi_{t} | \rho_{k}(t) | \chi_{t} \rangle = g_{\chi_{t}}^{\text{drift}}(k, t) + \langle \chi_{t} | \rho_{k}^{\text{eq}} | \chi_{t} \rangle \; P(k_{t}, k),$$  \hspace{1cm} (A4)$$
where the appearance of the factor $P(k_t, k)$ in the second term in Eq. (A4) accounts for the fraction of the initial equilibrium occupation that has survived the scattering.

The result Eq. (A4) is common to all situations (a), (b) and (c). If we are only interested in the steady states, we then take $\lim_{t \to -\infty} P(k_t, k) = 0$. The occupation on $(k_t, \chi_t)$ simply becomes

$$\langle \chi_t | \varrho_k (t) | \chi_t \rangle = \delta^{\text{drift}}_{k_t} (k_t, t).$$

(A5)

This is the starting point for the construction of the electron distribution function in the kinetic theory framework in Ref. [33] which is the basis also of Refs. [17, 30] for exploring Berry curvature effects under adiabatic and non-adiabatic conditions in the steady-state limits as the circumstances (a) and (b) mentioned above.

To make further progresses from Eq. (A5) (which amounts to further specifying the contents of Eq. (A3)), we need the assumption (ii) of the RTA. This assumption is based on the observation that in the absence of external fields, the collection of scattering events maintains the electronic system to obey some form of the distribution function, e.g., the Fermi-Dirac distribution function that describes the thermal equilibrium of the electron gases. Therefore, with the external field $E$ being turned on, we assume that there exists some form of distribution function $g^0_{k_t'} (k_t')$ such that whenever the electron occupation at a momentum $k_t'$ arrives this form, the occupation number should not be changed by the effects of scatterings. In other words, during the infinitesimal interval of length $d t'$ around time $t'$, the amount of electrons gained into the state $(k_t, \chi_t)$, denoted by $d g_{k_t'} (k_t', t')$, should equal to the amount of electrons lost from the occupation specified by $g^0_{k_t'} (k_t')$ due to scatterings, namely,

$$d g_{k_t'} (k_t', t') = \frac{d t'}{\tau_{k_t'}} g^0_{k_t'} (k_t').$$

(A6)

Further explicating $g^0_{k_t'} (k_t')$ then requires the differentiation between the circumstance (a) (under the adiabatic condition) and (b),(c) (not under the adiabatic condition).

a. case (a): adiabatic drivings

If the circumstance (a) is fulfilled, then an electron wave packet starting from one band $n$ at time $t'$ (carrying momentum $k_t'$) will remain at the same band $n$ under a different momentum $k_t$ at a later time $t > t'$. It is thus sufficient to consider $|\chi_{k_t'}\rangle = |u_n (k_{t'})\rangle$, and we denote $f_n (k_{t'}, t') := \langle \chi_{k_t'} | \varrho_k (t') | \chi_{k_{t'}} \rangle = |u_n (k_{t'})\rangle \rangle$ and $f_n^0 (k_{t'}) = g^0_{k_{t'}} (k_{t'})$ to follow the notational convention in the literatures. Here $f_n^0 (k_{t'}) = f^\mu_{FD} (\varepsilon_n (k_{t'}))$ describes the statistical mixture of the original bands $|u_n (k_{t'})\rangle$'s. Given the scattering rates $\tau_{k_{t'}}^{-1}$, the number of electrons lost/gained from/into the state $(k_{t'}, \chi_{k_{t'}}) = |u_n (k_{t'})\rangle$ for a differential time interval $d t'$ for Eq. (A6) is then given by

$$d g_{k_{t'}} (k_{t'}, t') = \frac{d t'}{\tau_{k_{t'}}} f_n (k_{t'}).$$

(A7)

Substituting Eq. (A7) into the right-hand side of Eq. (A3), and identifying $P(k_t, k_{t'}) / \tau_{k_{t'}} = \partial P(k_t, k_{t'}) / \partial t'$ (see [39]), then the integral over $t'$ there can be performed by using the integration-by-part technique. The result for Eq. (A5) with $f_n (k_t, t) = \langle \chi_t | \varrho_k (t) | \chi_t \rangle = |u_n (k_t)\rangle$ is then $f_n (k_t, t) = f_n^0 (k_t) + \delta f_n (k_t, t)$ as given by Eq. (16) of the main text, omitting the only band index $n$. The deviation from equilibrium appears more often in the literature in the form of $\delta f_n (k_t, t) = -\tau_{k_t} \left[ \partial f_n^0 (k_t) / \partial k_t \right] \cdot (-e/h) E (t)$ [18, 17, 38, 39, 53]. This is arrived by further making the short-relaxation-time approximation to Eq. (16) of the main text, namely,

$$P(k_t, k_{t'}) f (t') \approx e^{\frac{-t-t'}{\tau_{k_{t'}}}} f (t)$$

(A8)

for arbitrary function $f (t')$ [37].

b. case (b): non-adiabatic drivings by time-independent electric fields

If the adiabatic condition (a) is not fulfilled, then the external electric field can hybridise the bands as we come to the circumstance (b). We now review such situation but with $E (t) = E$ as a constant in time. The hybridised bands, $|u_n (k_{t'})\rangle$'s, each as a superposition of the original bands $|u_n (k_{t'})\rangle$'s, depend non-perturbatively on $E$ and are eigenfunctions of the moving-frame Hamiltonian $\tilde{H} (k_t, E)$ (see its definition below Eq. (18) in the main text) with
eigenenergies $\mathcal{E}_i(\mathbf{k}_i')$'s. The joint action of the decoherence and the electric field $E$ results in a statistical mixture of the hybridised bands, leading to

$$g_{\chi_i'}^0(\mathbf{k}_i') = \sum_i |\langle u_i(\mathbf{k}_i')|\chi_{i'}\rangle|^2 g_i^0(\mathbf{k}_i'), \quad (A9)$$

where $g_i^0(\mathbf{k}_i') = \int_{F_D}^T (\mathcal{E}_i(\mathbf{k}_i'))^{\mathbf{30,31}}$. This casts Eq. (A6) to

$$\hat{d}g_{\chi_i'}(\mathbf{k}_i', t') = \frac{dt'}{\tau_{k_{i'}}} \sum_i |\langle u_i(\mathbf{k}_i')|\chi_{i'}\rangle|^2 g_i^0(\mathbf{k}_i'). \quad (A10)$$

By applying similar procedures leading from Eq. (A7) to $f(\mathbf{k}_i, t)$ of Eq. (16) in the main text, additionally with Eq. (A8), here Eq. (A10) finally gives

$$g_i(\mathbf{k}_i, t) = g_i^0(\mathbf{k}_i) + \delta g_i(\mathbf{k}_i), \quad (A11)$$

where

$$\delta g_i(\mathbf{k}_i) = -\tau_{k_i} \frac{\partial g_i^0(\mathbf{k}_i)}{\partial k_{i'}} \left[ \frac{-e}{\hbar} \right] E, \quad (A12)$$

and we have notated $g_i(\mathbf{k}_i, t) = \langle u_i(\mathbf{k}_i)|g_k(t)|u_i(\mathbf{k}_i')\rangle$. How the result of Eq. (A11) reduces to the well-known result of (a) mentioned below Eq. (A7) in the limit of weak electric field has been detailed in Ref. [30]. Together with Eq. (4) (in the main text) for computing the currents, the main results of revealing the Berry curvature effects through the currents in Ref. [17] are also reproduced in Ref. [30] by taking the adiabatic limit.

Note that this steady-state non-adiabatic extension applied to gapped Dirac cone with two bands indexed as $c$ (conduction) and $v$ (valence) in 2D yields a non-adiabatically renormalised valley Hall current $j^H \propto E \times \int d^2k \mathbf{\Omega}(\mathbf{k}) / \sqrt{1 + 4 |r_{c,v}(\mathbf{k}, E)|}$ where $r_{c,v}(\mathbf{k}, E)$ is found in Eq. (12) of the main text [31]. It is non-perturbation in $E$ since $r_{c,v}(\mathbf{k}, E)$ appears in the denominator. However, the geometric manifestation is still through the Berry curvature $\mathbf{\Omega}(\mathbf{k})$ in this steady-state limit. By going to the transient regime with arbitrary time-dependent driving electric fields, it is possible to have geometric manifestation even with $\mathbf{\Omega}(\mathbf{k}) = 0$.

\textit{c. case (c): non-adiabatic drivings by time-dependent electric fields}

We are now facilitated to generalise the above construction (b) to the circumstance (c) with time-dependent electric field to obtain $\hat{g}_k(t)$ for $t$ not far away from $t_0$. Since the circumstance (b) is a special case of (c) by taking the electric field to be constant in time, we simply need to restore $E \rightarrow E(t')$ in $\hat{H}(\mathbf{k}_i', E(t'))$ for specifying $|u_i(\mathbf{k}_i')\rangle$ and $\mathcal{E}_i(\mathbf{k}_i')$ used in Eq. (A10) and substitute it to the right-hand side of Eq. (A3) to get

$$g_{\chi_i'}^{\text{drift}}(\mathbf{k}_i, t) = \int_{t_0}^t dt' \langle \chi_{i'}|\hat{g}_k(t')|\chi_{i'}\rangle \frac{\partial P(k_i, \mathbf{k}_i')}{\partial t'}, \quad (A13)$$

where $\hat{g}_k(t') = \sum_i g_i^0(\mathbf{k}_i')|u_i(\mathbf{k}_i')\rangle \langle u_i(\mathbf{k}_i')|$. (see also Eq. (18b) in the main text). Performing similarly the integral of Eq. (A13) using the integration-by-part method and applying $\langle \chi_{i'}\rangle = U_k(t', t)|\chi_i\rangle$ for $t_0 \leq t' \leq t$ to turn the right-hand side of Eq. (A4) to the form of $\langle \chi_{i'}|\bullet|\chi_i\rangle$ so that the density matrix $\hat{g}_k(t)$ can be identified with $\bullet$ in Eq. (A4), the result then is just Eq. (18) of the main text.

One can rewrite Eq. (18) of the main text via the integration-by-part method to

$$\hat{g}_k(t) = \hat{g}_k^{\text{ini}}(t) + \hat{g}_k^{\text{drif}}(t), \quad (A14a)$$

where

$$\hat{g}_k^{\text{ini}}(t) = U_k(t, t_0) \hat{g}_k^{\text{eq}} U_k(t_0, t) P(k_i, \mathbf{k}_i), \quad (A14b)$$

and

$$\hat{g}_k^{\text{drif}}(t) = \int_{t_0}^t dt' \frac{P(k_i, \mathbf{k}_i')}{\tau_{k_i'}} U_k(t, t') \hat{g}_k(t') U_k(t', t). \quad (A14c)$$

If we let $\tau_k \rightarrow \infty$, then $\int_{t_0}^t dt' \frac{\partial g_i^0}{\partial t'}(k_i', t'') P(k_i, \mathbf{k}_i') / \tau_{k_i'} \rightarrow 0$, $P(k_i, \mathbf{k}_i) \rightarrow 1$, and by the use of $\hat{g}_k(t_0) = \hat{g}_k^{\text{eq}}$, we have $g_k(t) \rightarrow \hat{g}_k^{\text{ini}}(t)$. So assuming the relaxation times to be infinite reduces the time-dependent density matrix of Eq. (18) to the intrinsic limit given by Eq. (10) in the main text (see also Eqs. (6) and (8) there).
3. Reproduction of Eq. (15) of the main text and second-order responses under periodic drivings

We should verify Eq. (18) by substituting it to Eq. (4) in the main text to obtain the current for arbitrary time-dependent electric field $E(t)$ and compare it to the known results in the literatures. We first show that under the adiabatic condition, such current is reduced to Eq. (15) of the main text, which has been applied to analyse non-perturbative intra-band effects (see references below Eq. (15) of the main text). If we further restrict the time-dependent electric fields to be time-periodic at times far away from $t_0$ and consider the current to the second order in the electric field, then it gives the Berry curvature dipole that serves as the basis of quantum nonlinear Hall effect [6].

Under the adiabatic condition, the weak applied field only weakly hybridises the bands. The index $i$ for labelling a hybridised band can thus be reduced to the label of an original band $n$, namely, $|u_n(k_i)⟩ ≈ e^{iγ_n(k_i)} |u_n(k_t)⟩ + |δu_n(k_i)⟩$, where $|δu_n(k_i)⟩$ is the perturbation to the first order in the electric field while to the same order $E_n(k_i) ≈ ε_n(k_i)$ so $j_0^i(k_i) ≈ J_0^i(k_i)$ reducing the first term of Eq. (18a) to

$$\tilde{ρ}_k(t) = \sum_n f_0^0(k_i)|u_n(k_i)⟩ ⟨u_n(k_i)|,$$  \hspace{1cm} (A15)

With the adiabatic condition imposed to the dynamics of a single wave packet, namely, $|u_n(k_i)⟩ = U_k(t, t') |u_n(k_i)⟩$, rendering $J^i_0(k_i, t, t') = \sum_n |u_n(k_i)⟩ ⟨u_n(k_i)| f_0^0(k_i)$ (see Eq. (18d)), the second term of Eq. (18) becomes

$$\delta J^i_0(t) = -\sum_n |u_n(k_i)⟩ ⟨u_n(k_i)| δJ_{nk}^{(1)}(t)$$ \hspace{1cm} (A16)

where

$$\delta J_{nk}^{(1)}(t) = \int_{t_0}^{t} dt' P(k_t, k_t') \frac{∂f_0^0(k_t)}{∂k_{t'}} \cdot \left(-\frac{e}{\hbar}\right) E(t'),$$ \hspace{1cm} (A17)

in which the term $\left(\frac{∂f_0^0(k_t)}{∂k_{t'}} \cdot (-e/\hbar) E(t')\right)$ comes from $df_n^0(k_{t'})/dt'$. The expectation values of the velocity operator (see its definition in Eq. (3) of the main text) on the weakly hybridised bands are given by,

$$⟨u_n(k_i)|V(k_i)|u_n(k_i)⟩ = v^b_n(k_i) + v^c_n(k_i, E(t)),$$ \hspace{1cm} (A18)

in which we have explained the meanings of $v^b_n(k_i)$ and $v^c_n(k_i, E(t))$ in the main text below Eq. (13) and one immediately identifies $\tilde{x}^a_n = ⟨u_n(k_i)|V(k_i)|u_n(k_i)⟩$. Taking the first line of Eq. (18) and putting it into the definition Eq. (4) in the main text, one obtains $j(t) = J(t) + \delta j(t)$ in which $J(t) = -e \int d^2k Tr(V(k_i) \tilde{ρ}_k(t))$ and $\delta j(t) = -e \int d^2k Tr(V(k_i) \delta ρ_k(t))$. Specifying $δ\tilde{ρ}_k(t)$ and $δ\tilde{ρ}_k(t)$ by Eqs. (A15) and (A16) respectively together with the use of Eq. (A18) on one band, one then straightforwardly obtains Eq. (15) of the main text.

We now turn to the second-order current. Note that $\tilde{j}(t)$ is only to the first order of $E(t)$ which gives the usual linear-response Hall current well reviewed in Ref. [17]. The second-order response then must be contained in $δj(t)$. One therefore has $δj(t) = δj^{(1)}(t) + δj^{(2)}(t)$ where $δj^{(1)}(t) = -e \int d^2k Tr(V(k_i) \delta ρ_k(t))$ is to the first order in the electric field and $δj^{(2)}(t) = -e \int d^2k Tr(V(k_i) \delta j_{nk}^{(1)}(t))$ accounts for the second-order response. We further assume that $τ_{k'} = τ$ (single-relaxation time) for all possible $k'$. We also assume that $τ$ is short so that Eq. (A8) can be applied. We only apply Eq. (A8) to the part $P(k_t, k_t') (δJ_{nk}^{(1)}(k_t)/\partial k_{t'})$ of the integrand in the integral of Eq. (A17), while the part of the integrand given by $E(t')$ is left intact [88]. We now impose the time-periodic driving $E(t') = Re\left\{E e^{iωt'}\right\}$ where $E$ is the time-independent complex electric-field amplitude and take the limit $t_0 \to -∞$, we then end up with

$$δj^{(2)}(t) = -\sum_{nα} \int d^2k f_n^0(k) \frac{∂Ω_n(k)}{∂k_α} × Re\left\{\frac{τ e^3}{2\hbar^2 (1 + iωτ)} \left(\tilde{E}_α\tilde{E}^* + e^{2iωτ} \tilde{E}_α\tilde{E}\right)\right\},$$ \hspace{1cm} (A19)

where the index $α$ runs over the spatial components and in the intermediate procedure, we have identified $d^2k$ with $d^2k_t$ and afterwards rewritten $k_t$ to $k$ for the integral. Here the Berry curvature $Ω_n(k)$ is given by Eq. (14) of the main text and the Berry curvature dipole is given by the components of $∂Ω_n(k)/∂k_α$. By focusing on one active band $n$, Eq. (A19) then reproduces the second-order response describing the quantum nonlinear Hall effect in accordance with Ref. [6].
4. The post-pulse current rates

In the above, we have derived the main formulation of NADT-WPT developed here as Eq. (18) of the main text and showed its reproductions of well known results of the AD-WPT. We now discuss the non-adiabatic transient consequences that one can find with such NADT-WPT. We first analyse the current rate \( \dot{j} (t) \) induced by a pulse of finite time width (subjecting to Eq. (20) of the main text) whose relation to manifesting geometric properties of the bands has been discussed in the main text Sec. II C 1. Taking the time derivative of Eq. (4) gives

\[
\dot{j}_\alpha (t) = \dot{j}^{\text{drv}}_\alpha (t) + \dot{j}^{\text{rp}}_\alpha (t),
\]

where

\[
\dot{j}^{\text{drv}}_\alpha (t) = \left( -e \right)^2 \hbar E (t) \cdot \int d^D k \text{Tr} \left( \frac{\partial V_\alpha (k)}{\partial k} \dot{\varrho}_k (t) \right),
\]

and

\[
\dot{j}^{\text{rp}}_\alpha (t) = -e \int d^D k \text{Tr} \left( V_\alpha (k) \dot{\varrho}_k (t) \right),
\]

in which \( \dot{j}_\alpha (t) \) and \( V_\alpha (k) \) are the \( \alpha \)th spatial component of \( \dot{j} (t) \) and \( V \) \( (k) \) respectively. Here the superscript “drv” for \( \dot{j}^{\text{drv}} \) indicates that this component instantaneously reflects the driving field \( E (t) \) and the superscript “rp” for \( \dot{j}^{\text{rp}} \) means that this component behaves in response to the driving field. Noticeably from setting \( E (t > t_{\text{off}}) = 0 \) to Eq. (A20), we see \( \dot{j}^{\text{drv}} (t > t_{\text{off}}) = 0 \). The post-pulse current rate then is only contributed by \( \dot{j} (t > t_{\text{off}}) = \dot{j}^{\text{rp}} (t) \).

We first clarify the intrinsic limit in which the scattering rates are zero \( \tau^{-1}_k \rightarrow 0 \) such that \( \dot{\varrho}_k (t) \rightarrow \dot{\varrho}_k^{\text{id}} (t) \) (see Eq. (10) of the main text). The extrinsic effects due to non-vanishing scattering rates will be discussed later. Taking the intrinsic limit of Eq. (18) leads to \( \dot{\varrho}_k (t) = (-i/\hbar) [H (k), \dot{\varrho}_k (t)] \) which in turn casts Eq. (A20c) to \( \dot{j}^{\text{rp}} (t) = \dot{j}^{\text{geo}} (t) \) as Eq. (21) of the main text, where quantum acceleration \( \dot{\kappa} (k) \), other than the anomalous velocity, appears and dominates the manifestation of geometric effects. We now explain why the manifestation of geometric properties of bands in the current rate after switching off the laser pulse is exclusive to non-adiabatic drivings but not adiabatic drivings during the in-pulse processes.

a. Exclusive effects of non-adiabatic drivings on post-pulse wave-packet dynamics

For adiabatic dynamics of a single wave packet, the expectation value \( \langle \phi^{\text{id}}_{n,k} (t) | O | \phi^{\text{id}}_{n,k} (t) \rangle \) of any observable \( O \) at any moment of time \( t \) depends on \( t \) only through \( k \). As the driving ceases for \( t > t_{\text{off}} \), Eq. (3) of the main text dictates that \( k \) no longer changes with time, namely, \( k_{\text{i}} | t > t_{\text{off}} = k + K_{\text{i,off}} \equiv k_{\text{off}} \). The expectation value also stops to change with time so its time derivative remains zero after the driving is switched off which corresponds to \( \dot{j} (t > t_{\text{off}}) = 0 \) in the above discussion.

In contrast, non-adiabatically, the time-dependent expectation value \( \langle \phi_{n,k} (t, t_{0}) | O | \phi_{n,k} (t, t_{0}) \rangle \) of an arbitrary observable \( O \) depends on \( t \) through the whole driving history up to time \( t \) more than just \( k \) at that moment. As the non-adiabatic drivings enable \( | \phi_{n,k} (t_{\text{off}}, t_{0}) \rangle \) to acquire non-negligible occupations on bands other than the starting band \( n \), the state \( | \phi_{n,k} (t_{\text{off}}, t_{0}) \rangle \) becomes a driving-history dependent superposition of eigenstates of \( H (k_{\text{off}}) \) as a genuine non-adiabatic multiple-band effect. Consequently, as the wave packet continues to evolve to the post-pulse time region as \( | \phi_{n,k} (t > t_{\text{off}}, t_{0}) \rangle \rightarrow \exp \left( -i/\hbar \right) (t - t_{\text{off}}) H (k_{\text{off}}) | \phi_{n,k} (t_{\text{off}}, t_{0}) \rangle \), the time derivative of the observables, e.g., the quantum acceleration, \( \dot{\kappa} (k) \), for \( t > t_{\text{off}} \) does not vanish.

Explicitly, such non-adiabatic effects from transient drivings \( E (t_0 \leq t \leq t_{\text{off}}) \neq 0 \) can be explicated in the post-pulse time region \( t > t_{\text{off}} \) in which \( E (t > t_{\text{off}}) = 0 \) from two complementary views. First, with a fixed pulse whose peak amplitude is large enough to activate non-adiabatic regime, two different band structures with the same band gaps but differentiated only by their geometric connections can leave such geometric differences imprinted to the band-pseudospin state \( | \phi_{n,k} (t_{\text{off}}, t_{0}) \rangle \) at the end of the pulse and manifested through the driving-free dynamics governed by \( H (k_{\text{off}}) \) in the post-pulse time region. Second, two different pulses are expected to induce different in-pulse dynamics as usual. Therefore, the post-pulse differences due to different pulses applied before are then evidences of history dependencies accumulated to \( | \phi_{n,k} (t_{\text{off}}, t_{0}) \rangle \) by non-adiabatic in-pulse drivings. These microscopic effects on the dynamics of the band pseudospin of a wave packet is then manifested in \( \dot{j} (t > t_{\text{off}}) \) as macroscopic observable through the quantum acceleration mechanism as discussed around Eqs. (21) and (22) in the main text.
b. Inclusion of extrinsic scattering effects

We now conclude the general discussion of post-pulse current rate by taking into account the extrinsic scattering effects. In the post-pulse time region, we always have \( \dot{j}^{drv} (t > t_{off}) = 0 \) due to its direct proportionality to the electric field at time \( t \) as Eq. (A20b) regardless of whether the scattering effects are ignored. For convenience of analysing the post-pulse current, instead of using Eq. (18) for the density matrix, here we substitute its alternative expression Eq. (A14) to Eq. (A20c) and obtain

\[
\dot{j}^{rp} (t) = \dot{j}^{rp,ini} (t) + \dot{j}^{rp,drf} (t),
\]

where \( \dot{j}^{rp,ini} (t) = -e \int d^Dk \text{Tr} \left( V (k) \dot{\varrho}_k (t) \right) \) and \( \dot{j}^{rp,drf} (t) = -e \int d^Dk \text{Tr} \left( V (k) \dot{\varrho}_k (t) \right) \) are the \( \alpha \) component of the vectors \( \dot{j}^{rp,ini} (t) \) and \( \dot{j}^{rp,drf} (t) \) respectively. More explicitly,

\[
\dot{j}^{rp,ini} (t) = \dot{j}^{vel,ini} (t) + \dot{j}^{acc,ini} (t),
\]

where

\[
\dot{j}^{vel,ini} (t) = -e \int d^Dk \left[ \frac{P (k_{1}, k)}{\tau_k} \right] \text{Tr} \left( V (k) \dot{\varrho}_k (t) \right),
\]

\[
\dot{j}^{acc,ini} (t) = -e \int d^Dk P (k) \text{Tr} \left( \hat{A} (k) \dot{\varrho}_k (t) \right),
\]

in which \( \dot{\varrho}_k (t) \) is found from Eqs. (10) and (8), \( V (k) \) is defined in Eq. (8) and \( \hat{A} (k) \) is still the quantum acceleration defined in Eq. (21b) in the main text respectively.

To take the time derivative of \( \dot{\varrho}_k (t) \) to obtain \( \dot{j}^{rp,drf} (t) \), we first note that there exists a time scale \( \tau_m \geq \max \{ \tau_k \} \) such that \( P (k_{1}, k_{2}) \approx 0 \) for \( t - t' \geq \tau_m \) so the integral \( \int_{t_0}^{t} dt' \) in Eq. (14c) is only contributed by its integrand at \( t' \) that satisfies \( t - \tau_m \leq t' \leq t \). By further realising that \( \dot{\varrho}_k (t' > t_{off}) = \varrho_k^{eq} \) and \( U_k (t, t' > t_{off}) = \exp \left( \frac{-i}{\hbar} (t - t') \mathcal{H} (k_{t_{off}}) \right) \) so \( \dot{\varrho}_k (t') = \varrho_k^{eq} \) \( \forall t' > t_{off} \) in Eq. (14c), it becomes

\[
\dot{\varrho}_k^{drf} (t > t_{off} + \tau_m) \approx \left[ \int_{t - \tau_m}^{t} dt' P (k_{1}, k_{2}) / \tau_{k_{t'}} \right] \varrho_k^{eq}.
\]

The integral over \( t' \) is found to be \( \int_{t - \tau_m}^{t} dt' P (k_{1}, k_{2}) / \tau_{k_{t'}} = \int_{t - \tau_m}^{t} dt' \partial P (k_{1}, k_{2}) / \partial t' = 1 - P (k_{1}, k_{2} - \tau_m) \) by the use of integration by part. Noting for \( t \geq t' > t_{off} \), one has \( P (k_{1}, k_{2}) = \exp \left\{ - \int_{t'}^{t} dt'' \tau_{k_{t''}} \right\} = \exp \left\{ - \int_{t'}^{t} dt'' \tau_{k_{t''}} \right\} = \exp \left\{ - (t - t') / \tau_{k_{t''}} \right\} \). Therefore, \( P (k_{1}, k_{2} - \tau_m) = \exp \left\{ - (t - t') / \tau_{k_{t''}} \right\} \) which then gives \( \dot{\varrho}_k^{drf} (t > t_{off} + \tau_m) \approx \left[ 1 - \exp \left\{ - \tau_{m} / \tau_{k_{t''}} \right\} \right] \varrho_k^{eq} \) leading to \( \dot{\varrho}_k^{drf} (t > t_{off} + \tau_m) = 0 \) and \( \dot{j}^{rp,drf} (t > t_{off} + \tau_m) = 0 \). Consequently, by Eq. (A21) with \( \dot{j}^{rp,drf} (t > t_{off} + \tau_m) = 0 \), we obtain

\[
\dot{j} (t > t_{off} + \tau_m) \approx \dot{j}^{rp,ini} (t),
\]

where \( \dot{j}^{rp,ini} (t) \) has been found as Eq. (A22). Comparing Eq. (A22c) for \( \dot{j}^{acc,ini} (t) \) with Eq. (21) of the main text for \( \dot{j}^{geo} (t) \), we see that the post-pulse current rate \( \dot{j} (t > t_{off} + \tau_m) \) given by Eq. (A24) still retain the manifestation of geometric effects through the quantum acceleration, albeit it is damped by the factor \( 0 < P (k_{1}, k) < 1 \) due to scattering. If adiabatic drivings had been applied in the time region \( t_0 \leq t \leq t_{off} \), then \( \dot{j}^{acc,ini} (t > t_{off}) = 0 \) by the reason that makes \( \dot{j}^{geo} (t) = 0 \) pointed out above. For longer relaxation times, both the damping factor \( P (k_{1}, k) \) and the contamination of this manifestation by \( \dot{j}^{vel,ini} (t) \) can be reduced.

Appendix B: Wave-packet spin dynamics and SOC-geometric effects

In Secs. IIC2 and IIC3 of the main text, we have used the concept that driving-induced intrinsic spin coherence represents SOC geometric effects to illustrate the prospects of macroscopically manifesting microscopic SOC-geometric
effects through CISP and spin-mediated part of the photocurrent. Here in Sec. B1 we explain this basic concept from the wave packet point of view. With this conceptual picture in mind, we then continue in Sec. B2 to study characteristics of driven spin-texture dynamics in the post-pulse time region. This also explicates the first abstract point about transient non-adiabatic manifestation of band-geometric effects discussed in Sec. H1C of the main text that relates such effects embedded in the quantum acceleration to the post-pulse current rate. In Sec. B3 we recall the well-established AD-WPT applied to CISP effects to provide relevant reference results to be compared with non-adiabatic CISP effects addressed in Secs. H1C2 and H1B of the main text.

1. Spin geometric connections for intrinsic spin coherences

We now inspect dynamics of the spin DOF of the wave packet under external drivings with the general Hamiltonian given by Eq. (23) of the main text. Let us consider a driving that pushes the CM of the wave packet in the BZ from $k_{t_0} = k$ at time $t_0$ to $k_t$ at time $t$ along a path $\Gamma_{t_0}$. We have introduced in Sec. H1C2 of the main text the driven spin texture $\langle \sigma \rangle_n(k, t)$ which includes effects of drivings so that $\langle \sigma \rangle_n(k, t) = \langle \phi_{n,k}(t, t_0) | \sigma | \phi_{n,k}(t, t_0) \rangle$ is expected to deviate from $\langle \sigma \rangle_n^0(k_t)$ (the native spin texture, defined by Eq. (25) in the main text). Consequently, the spin DOF of the wave packet described by the state $| \phi_{n,k}(t, t_0) \rangle$ becomes a coherent superposition between $|u_-(k_t)\rangle$ and $|u_+(k_t)\rangle$ showing intrinsic spin coherence. The native spin texture along the path $\Gamma_{t_0}$ determines how such driving induction of the intrinsic spin coherence can be incurred. Below we explain this for two opposite situations, namely, $\partial \langle \sigma \rangle_n^0(k_{t'}) / \partial k_{t'} \neq 0$ and $\partial \langle \sigma \rangle_n^0(k_{t'}) / \partial k_{t'} = 0$ for $t_0 \leq t' \leq t$.

We first consider a path that fulfills $\partial \langle \sigma \rangle_n^0(k_{t'}) / \partial k_{t'} \neq 0$ for $t_0 \leq t' \leq t$ (indicated by Eq. (26) of the main text). For general non-adiabatic drivings, we have $(\hbar/2) \langle \sigma \rangle_n(k, t) = \Lambda_{so}(k_t) \times \langle \sigma \rangle_n(k, t)$. This describes the spin precession dynamics in an effective time-dependent magnetic field $\Lambda_{so}(k_t)$ whose orientation is determined by the native spin texture $\Lambda_{so}(k_t) / |\Lambda_{so}(k_t)| = \pm \langle \sigma \rangle_n^0(k_t)$. The driving of the CM of the wave packet in the BZ along the path $\Gamma_{t_0}$ characterised by $\partial \langle \sigma \rangle_n^0(k_{t'}) / \partial k_{t'} \neq 0$ for $k_{t'} \in \Gamma_{t_0}$ keeps the orientation of the effective magnetic field $\Lambda_{so}(k_t)$ changing in time which causes the driven spin texture $\langle \sigma \rangle_n(k, t)$ to deviate from $\langle \sigma \rangle_n^0(k_t)$. This thus arrives a coherent superposition between $|u_-(k_t)\rangle$ and $|u_+(k_t)\rangle$ for the spin DOF of the wave packet. In case of adiabatic drivings, the spin state of the wave packet is simplified to $| \phi_{n,k}(t, t_0) \rangle \rightarrow | \phi_{n,k}^0(t) \rangle$ given by Eq. (11) of the main text which obviously shows spin coherence at $k_t$ as superposition between $|u_-(k_t)\rangle$ and $|u_+(k_t)\rangle$ because $r_{n,n}(k_t, E(t)) \neq 0$ by $|\mathcal{R}_{k_{t'}}\rangle_{n,n} \neq 0$. The deviation of $\langle \sigma \rangle_n(k, t)$ from $\langle \sigma \rangle_n^0(k_t)$ is on the first order of the driving field $E(t)$. Therefore, regardless whether the driving is adiabatic or non-adiabatic, driving-induced intrinsic spin coherence is attained due to the effects of the non-vanishing inter-spin geometric connection $|\mathcal{R}_{k_{t'}}\rangle_{n,n} \neq 0$ associated with the native spin texture $\partial \langle \sigma \rangle_n^0(k_{t'}) / \partial k_{t'} \neq 0$. Such driving-induced intrinsic spin coherence thus faithfully represents the SOC-geometric effects.

We now consider the opposite case that the native spin texture along the path $\Gamma_{t_0}$ instead fulfills $\partial \langle \sigma \rangle_n^0(k_{t'}) / \partial k_{t'} = 0$ for $t_0 \leq t' \leq t$. This implies that the direction of $\Lambda_{so}(k_{t'})$ is a time constant given by $\langle \sigma \rangle_n^0(k_{t_0}) = k_t$. With the initial value of the driven spin texture set by $\langle \sigma \rangle_n(k, t_0) = \langle \sigma \rangle_n^0(k)$, we have $\langle \sigma \rangle_n(k, t') \mid_{t'=t_0} \propto \langle \sigma \rangle_n^0(k) \times \langle \sigma \rangle_n(k, t_0) = 0$ such that $\langle \sigma \rangle_n(k, t_0 + \delta t) = \langle \sigma \rangle_n(k, t_0)$ for small enough $\delta t > 0$. This can be iterated throughout the path $\Gamma_{t_0}$ to see that $\langle \sigma \rangle_n(k, t_0 \leq t' \leq t) = \langle \sigma \rangle_n(k, t_0) = \langle \sigma \rangle_n^0(k)$. Along such path on which the native spin texture fulfills $\partial \langle \sigma \rangle_n^0(k') / \partial k' = 0$ for $k' \in \Gamma_{t_0}$, the CM of the wave packet and its spin are essentially decoupled so the geometric properties of the bands are not manifested in wave-packet dynamics even if the inter-spin connections can be nonzero elsewhere. This is why the persistent spin texture, characterised by $\partial \langle \sigma \rangle_n^0(k') / \partial k' = 0$, excludes itself from our interests. In contrast, for the Rashba-Dresselhaus spin texture, one easily finds driving paths $\Gamma_{t_0}$ endowed with the geometric property $\partial \langle \sigma \rangle_n^0(k') / \partial k' \neq 0$ for $k' \in \Gamma_{t_0}$ that allows driving-induced intrinsic spin coherence.

2. Post-pulse driven spin texture dynamics linked to current rate

We have explained that the driven spin texture can show the intrinsic spin coherence induced by drivings to manifest underlying SOC-geometric effects. Such microscopic manifestation goes without distinguishing non-adiabatic and adiabatic drivings. We have discussed in Sec. H1C of the main text that the geometric manifestation of the non-adiabatic drivings can be exclusively captured in the post-pulse time region by the current rate as a macroscopic observable. We therefore should link the microscopic driven spin texture to the macroscopic current rate.
With the general Hamiltonian given by Eq. (23) of the main text, the quantum acceleration given by Eq. (22) of the main text reads

$$ \mathcal{H}_\alpha (k_i) = 2\hbar^{-2} \left[ \frac{\partial \Lambda_{so} (k_i)}{\partial k_{\alpha t}} \times \Lambda_{so} (k_i) \right] \cdot \sigma, \quad (B1) $$

and consequently

$$ j_{\alpha}^{geo} (t) = 2 (-e) \hbar^{-2} \int d^3k \left[ \frac{\partial \Lambda_{so} (k_i)}{\partial k_{\alpha t}} \times \Lambda_{so} (k_i) \right] \cdot (\sigma) (k, t), \quad (B2) $$

where $k_{\alpha t}$ denotes the $\alpha$th component of $k_i$. The above discussed geometric effects realised through the driven (and therefore the non-equilibrium) spin texture, namely, $(\sigma) (k, t) = \sum_n \int_{F \bar{F}} (\varepsilon_n (k)) (\sigma) (k, t)$, is then concretely manifested in the post-pulse current rate Eq. (B2) that does not vanish only for non-adiabatic driving during the pulse. Below we will discuss the properties of the dynamics of the driven spin texture $(\sigma)_n (k, t)$ in more detail.

In Sec. IIIA of the main text, we have noted that the SSH system is obtained by simply replacing the unspecified $\Lambda_{so} (k)$ by $\Lambda_{SSH} (k)$ specified in the main text. Therefore, by corresponding the spin to the sublattice pseudospin, the discussions below about the properties of the driven spin texture and their implications for the current rates are also valid for the SSH system as illustrated in Sec. IIIA of the main text.

To better understand the non-adiabatic driving effects on the spin texture, we consider a pulsing scenario in which $K_i |_{t > t_{off}} \rightarrow 0$ in Eq. (7) of the main text. We find it convenient to define $(\sigma)_{n,m} (k) := \langle u_n (k) | \sigma | u_m (k) \rangle$. Noting $(\sigma)_{n,n} (k) = \langle \sigma \rangle_n (k)$, the diagonals/off-diagonals of $(\sigma)_{n,m} (k)$ are called the native intra-band/inter-band spin texture. As $k_{off} \rightarrow k$, the driven spin texture in the post-pulse time region is universally given by

$$ (\sigma)_{n} (k, t > t_{off}) = (\sigma)_{n,dc}^{off} (k) + (\sigma)_{n,ac}^{off} (k, t). \quad (B3a) $$

It contains a time-independent contribution, $(\sigma)_{n,dc}^{off} (k) = \sum_m |\varphi_{mk} (t_{off})|^2 (\sigma)_{m}^{0} (k)$, in which $\varphi_{mk} (t) = \langle u_m (k) | \phi_n (k, t, t_0) \rangle$ and a time-oscillating contribution,

$$ (\sigma)_{n,ac}^{off} (k, t) = \sum_m \sum_{m' \neq m} e^{-(i/\hbar)(t-t_{off})(\varepsilon_m - \varepsilon_{m'}) (k)} \varphi_{m'k}^{*} (t_{off}) (\sigma)_{m'}^{0} (k = \langle \sigma \rangle_{m',m} (\varphi_{mk} (t, t_0). \quad (B3b) $$

In Eq. (B3), we thus see clearly how the different band-structure factors (spin texture and band gaps) and the external-driving scenarios $E (t_0 \leq t \leq t_{off})$ come together to determine the behaviours of the dc and the ac components of the driven spin texture in the post-pulse time region. The dc component $(\sigma)_{n,dc}^{off} (k)$ is solely characterised by its magnitude as a time-independent offset. The dynamics of the ac component is characterised by three attributes, namely, the amplitude, the phase and the period of the ac oscillation. The result of Eq. (B3) says that the native intra-band spin texture $(\sigma)_{m}^{0} (k)$ determines the magnitudes of the dc component while the native inter-band spin texture $(\sigma)_{m',m} (k)$ is one factor behind the amplitude of the ac component. The band gaps $(\varepsilon_m - \varepsilon_{m'}) (k)$ determine the periods of the ac oscillations in time.

We now expound on the interplay between the in-pulse non-adiabatic drivings and the geometric properties of the bands. We address how such interplay manifests itself in the post-pulse driven spin texture dynamics and the current rate in correspondence to the points (i) and (ii) abstracted in Sec. IIIA of the main text. Certainly, the band occupation $|\eta_{mk} (t_{off})|^2$ and inter-band coherence $\eta_{m'k}^{*} (t_{off}) \eta_{mk} (t_{off})$ of the wave-packet band pseudospin at the end of the pulse are results of CM-band-pseudospin coupled dynamics that combined effects from both the band-geometric properties and the drivings as explained before. Here, more specifically for the post-pulse driven spin textures, the band occupation and the inter-band coherence at the end of the pulse respectively affect the magnitudes of the dc and the amplitudes of the ac components as told by Eq. (B3). This illustrates the point (i) about how the in-pulse drivings allow the geometric properties of the bands to leave imprints on the post-pulse current rate $\hat{j} (t > t_{off}) = \hat{j}^{geo} (t)$. As indicated by Eq. (B2), such imprints are realised via the underlying driven spin texture $(\sigma) (k, t > t_{off})$.

For the point (ii), we should consider two different non-adiabatic driving scenarios $A$ and $B$ to inspect the geometric significance of history-dependence effects. Different in-pulse drivings are not expected to result in different periods of the post-pulse ac oscillations because the period is determined by the band gaps $(\varepsilon_m - \varepsilon_{m'}) (k)$, in accordance to Eq. (B3). Effects of different driving scenarios can be better envisaged by assuming that the two pulses have the same duration $t_{off} - t_0$ but leading to different inter-band coherence at the end of the pulse, namely, $\eta_{m'k}^{*} (t_{off}) \eta_{mk} (t_{off}) |_{A} \neq \eta_{m'k}^{*} (t_{off}) \eta_{mk} (t_{off}) |_{B}$. We now take the time $t$ in Eq. (B2) to be in the post-pulse time region, namely, $j_{\alpha}^{geo} (t > t_{off}) = 2 (-e) \hbar^{-2} \int d^3k \left[ \frac{\partial \Lambda_{so} (k) / \partial k_{\alpha t}}{\partial k_{\alpha t}} \times \Lambda_{so} (k) \cdot (\sigma) (k, t > t_{off}).
Here we have applied \( \Lambda_{so}(k_i)|_{t>t_{off}} = \Lambda_{so}(k_{t_{off}}) \rightarrow \Lambda_{so}(k) \). In terms of the microscopic driven spin texture, it is clear that \( \langle \sigma \rangle (k, t > t_{off})|_A \) and \( \langle \sigma \rangle (k, t > t_{off})|_B \) share the same oscillation period while the situation \( \eta_{mk}(t_{off}) \eta_{mk}(t_{off})|_A \neq \eta_{mk}(t_{off}) \eta_{mk}(t_{off})|_B \) caused by different drivings only result in differences in the ac oscillation amplitudes between \( \langle \sigma \rangle (k, t > t_{off})|_A \) and \( \langle \sigma \rangle (k, t > t_{off})|_B \), as already discussed with Eq. (B3) above. Macroscopically, however, for the current rate, the above microscopic \( k \)-dependent amplitude difference can give rise to different oscillation phases between \( j^{geo}_\alpha(t > t_{off})|_A \) and \( j^{geo}_\alpha(t > t_{off})|_B \) after the integration over \( k \). Therefore, different in-pulse non-adiabatic drivings can give rise to different oscillation phases rooted to the band-geometric effects.

3. Properties of adiabatic CISP

We have discussed the motivations of studying CISP from the perspective of SOC-geometric effects in Sec. IIIC2 of the main text. With Rashba SOC, we have explored their salient transient non-adiabatic manifestations in Sec. IIIB of the main text. Here we supplement information about CISP under adiabatic drivings first without specifying the SOC to be Rashba. This readily allows one to see the cancellation of the intrinsic CISP. We then specify to the setting of Rashba SOC to investigate properties of the remained CISP effects involving extrinsic mechanisms more explicitly.

a. Cancellation of intrinsic spin coherence effects in CISP

Based on Eq. (23) of the main text, we first recap that the adiabatically induced CISP in the linear response regime does not retain properties of the intrinsic spin coherence in the resulting macroscopic spin polarisation. This goes without any specification of the spin-orbit field \( \Lambda_{so}(k) \) except that it satisfies

\[
\Lambda_{so}(k) = -\Lambda_{so}(-k),
\]

as a result of respecting TRS.

In the adiabatic regime to the first order in \( E(t) \), the driven spin texture given by \( \langle \sigma \rangle^{ad}_n(k, t) := \left\langle \phi^{ad}_n(k, t) \right| \sigma \left| \phi^{ad}_n(k, t) \right\rangle \) with \( \phi^{ad}_n(k, t) \) found in Eq. (11) of the main text leads to

\[
\langle \sigma \rangle_n^{ad}(k, t) = \langle \sigma \rangle_n^0(k_i) + \langle \sigma \rangle_n^{r}(k_i, E(t))
\]

(B5a)

where \( \langle \sigma \rangle_n^{r}(k_i, E(t)) \) is linear in \( E(t) \), i.e.,

\[
\langle \sigma \rangle_n^{r}(k_i, E(t)) = \sum_\alpha \chi^{\alpha}_\beta(k_i)(-cE_\alpha(t)),
\]

(B5b)

with linear-response coefficients:

\[
\chi^{\alpha}_\beta(k_i) = \frac{\langle u^\alpha_n(k_i) | \sigma_\beta | u_n(k_i) \rangle [R_{\alpha,n,n}]_{\beta,n}}{\varepsilon_\beta(k_i) - \varepsilon_\alpha(k_i)} + \text{c.c.},
\]

(B5c)

where \( \tilde{n} \) stands for the spin opposite of \( n \), e.g., \( n = + \) then \( \tilde{n} = - \) and vice versa. Here in Eq. (B5), \( \langle \sigma \rangle_n^{r}(k_i, E(t)) \) represents the part of the driven spin texture that contains effects from the adiabatically-induced intrinsic spin coherence. As shown in Eq. (B5c), this is enabled by \( [R_{\alpha,n,n}]_{\beta,n} \neq 0 \) as a geometric effect. Without restricting the driving field to the adiabatic regime, the driven spin texture \( \langle \sigma \rangle_n(k, t) \) in general develops complicated time-dependent field configurations over the domain of \( k \). In contrast, under the adiabatic limit, \( \langle \sigma \rangle_n(k, t) \rightarrow \langle \sigma \rangle_n^{ad}(k, t) \), the description of the spin-texture field is simplified to Eq. (B5) in which the time development of \( E(t) \) is factored out. Effectively, the driven spin texture is characterised by \( \chi^{\alpha}_\beta(k_i) \)'s as static field configurations over the domain of \( k_i \). By the TRS of Eq. (B4), one sees that \( \chi^{\alpha}_\beta(k_i) = -\chi^{\alpha}_\beta(-k_i) \) and therefore

\[
\langle \sigma \rangle_n^{r}(k_i, E(t)) = -\langle \sigma \rangle_n^{r}(k_i, E(t)).
\]

(B6)

This similarity to the anti-symmetric character of the native spin texture is a signature of the SOC-geometric effects during adiabatic driving processes.

We can calculate the non-equilibrium spin polarisation due to the adiabatic drivings in the linear response regime by fully ignoring band hybridisation in Eq. (18) for the density matrix (so \( \tilde{\gamma}_k(t) = \sum_n f_n^0(k_i) | u_n(k_i) \rangle \langle u_n(k_i) | \) but
maintaining the non-equilibrium deviation Eq. \(A_{16}\) caused by the external field. We further make the approximation given by Eq. \(A_{8}\). The resulting density matrix substituted to Eq. \(B_{29}\) (of the main text) as the definition for spin polarisation (here renotated to \(\hat{\mathbf{S}}(t) \rightarrow \hat{\mathbf{S}}^{ad}(t)\) for adiabatic drivings) then gives

\[
\hat{\mathbf{S}}^{ad}(t) = \hat{\mathbf{S}}(t) + \delta \hat{\mathbf{S}}(t),
\]

(B7a)

where

\[
\hat{\mathbf{S}}(t) = \frac{\hbar}{2} \sum_{n} \int d^{D} \mathbf{k} \langle \sigma \rangle^{\tau}_{n}(\mathbf{k}, E(t)) f^{0}_{n}(\mathbf{k}),
\]

(B7b)

and

\[
\delta \hat{\mathbf{S}}(t) = -\tau \frac{\hbar}{2} \sum_{n} \int d^{D} \mathbf{k} \langle \sigma \rangle^{0}_{n}(\mathbf{k}) \left[ \frac{\partial f^{0}_{n}(\mathbf{k})}{\partial \varepsilon_{n}(\mathbf{k})} \right] (-e) E(t),
\]

(B7c)

in which we have used \(d^{D} \mathbf{k} = d^{D} \mathbf{k}\). The intrinsic spin-split-bands’ geometric effects appear in the macroscopic spin polarisation via \(\hat{\mathbf{S}}(t)\) while \(\delta \hat{\mathbf{S}}(t)\) in proportion to the relaxation time \(\tau\) is coupled with extrinsic scattering. By Eq. \(B_{6}\) and noting \(f^{0}_{n}(\mathbf{k}) = f^{0}_{n}(-\mathbf{k})\) due to the Kramer degeneracy given by the TRS, we have

\[
\hat{\mathbf{S}}(t) = 0.
\]

(B8)

The adiabatic driving thus limits the nonzero spin polarisation arising from the CISP effect to contain only the extrinsic contribution \(\hat{\mathbf{S}}^{ad}(t) = \delta \hat{\mathbf{S}}(t)\), reproducing the results in \[72\] [73].

b. Restrictions on the CISP orientation for Rashba SOC under adiabatic drivings

We now specify \(\Lambda_{so}(\mathbf{k})\) to be the spin-orbit field for the anisotropic Rashba SOC defined in Eq. \(34\) of the main text. Under adiabatic drivings, the driven spin texture from Eq. \(B_{5}\) becomes

\[
\langle \sigma \rangle^{\tau}_{n}(\mathbf{k}, E(t)) = \left( \frac{\hat{\mathbf{z}} \times \mathbf{k}}{2 \alpha R |\chi_{k}^{F} \mathbf{k}|} \right) (-e) E(t),
\]

(B9)

while \(\langle \sigma \rangle^{0}_{n}(\mathbf{k}, E(t)) = 0\). The antisymmetry of \(\langle \sigma \rangle^{\tau}_{n}(\mathbf{k}, E(t))\) in \(\mathbf{k}\) (as discussed with Eq. \(B_{6}\)) is made explicit here by Eq. \(B_{9}\). We already know from the discussion for Eq. \(B_{8}\) that under the adiabatic driving, the CISP is solely produced by the extrinsic contribution \(\hat{\mathbf{S}}^{ad}(t) = \delta \hat{\mathbf{S}}(t)\). By the nature of Eq. \(34\) of the Rashba spin-orbit field, the extrinsic CISP given by \(\delta \hat{\mathbf{S}}(t)\) of Eq. \(B_{7c}\), in which \(\langle \sigma \rangle^{0}_{n}(\mathbf{k}) \cdot \hat{\mathbf{z}} = 0\), is limited to be in-plane which in turn supports that an out-of-plane CISP should reveal the intrinsic properties. Under the isotropic condition, namely, \(r_{A} = 1\), the native spin texture in Eq. \(B_{7c}\) becomes \(\langle \sigma \rangle^{0}_{n}(\mathbf{k}) = \pm \mathbf{k} \times \hat{\mathbf{z}}/|\mathbf{k}|\). As a result, the orientation of the CISP is simplified to \(\hat{\mathbf{S}}^{ad}(t) \parallel \hat{\mathbf{e}} \times \hat{\mathbf{z}}\), as also found in \[72\] [73]. Therefore the adiabatic driving to the linear response regime limits the spin polarisation orientation raised by the CISP effect to

\[
\hat{\mathbf{z}} \cdot \hat{\mathbf{S}}^{ad}(t) = 0,
\]

(B10)

\[
\hat{\mathbf{S}}^{ad}(t) \cdot \hat{\mathbf{e}}|_{r_{A}=1} = 0.
\]

(B11)

Even though the intrinsic spin coherence evidenced by Eq. \(B_{9}\) is not manifested through CISP, it has been shown to be manifested through the spin Hall current (not the ordinary electronic current Eq. \(4\) defined in the main text) and serves as the basis of the universal intrinsic spin Hall effect (see Ref. \[50\] in which the isotropic limit \(\chi_{k} \mathbf{k} = \mathbf{k}\) is considered). We will show later that the intrinsic spin coherence effect induced by non-adiabatic drivings can also be manifested directly in the electronic current and in the CISP under appropriate circumstances (see discussions below).

Appendix C: Supporting information on the Rashba-SOC system

In this section, we supplement information on the spin polarisation and photocurrent dynamics to support the statements about the anisotropic Rashba system briefed in Sec. \[B18\] of the main text.
1. Non-adiabatic dynamics of driven spin texture and CISP

In the non-adiabatic regime, to more conveniently study the driven spin texture, which is generally asymmetric, we define \( \langle \sigma \rangle (k, t) := \langle \sigma \rangle_{n_+}(k, t) + \langle \sigma \rangle_{n_-}(-k, t) \) by the virtue that before the electric field is turned on it is zero, namely, \( \langle \sigma \rangle (k, t_0) = 0 \) for all \( k \)'s, due to the TRS of the native spin texture. Here \( n_- \) labels the lower energy band for a given \( k \) and \( (-n_-, -k) \) labels the Kramer partner of \( (n_+, k) \). We assume the initial equilibrium is given by filling up only the lower-energy band. The intrinsic spin polarisation is then given by \( S \) labels the lower energy band

\[
\langle \sigma \rangle_{n_{\pm}}(k, t) = \delta s / G_0 (\theta, \varphi_e), \quad \text{for} \quad \theta \leq \pi/2,
\]

where

\[
G_0 (\theta, \varphi_e) = 2 \bar{r} (\cos [2 (\theta - \varphi_e)] - 1) + \delta r (\cos (2\theta) - \cos (2\varphi_e)) / \sqrt{r_X^2 \cos^2 \theta + r_Y^2 \sin^2 \theta},
\]

and

\[
G_1 (x, \theta, \varphi_e) = - (x + x^{-1}) \sin [2 (\theta - \varphi_e)] + (x - x^{-1}) [\sin (2\varphi_e) - \sin (2\theta)] / \sqrt{r_X^2 \cos^2 \theta + r_Y^2 \sin^2 \theta},
\]

in which

\[
\bar{r} = (r_Y + r_X) / 2,
\]

\[
\delta r = (r_Y - r_X) / 2.
\]

Recall from the main text that \( r_X = \sqrt{m_y / m_z}, \) \( r_Y = \sqrt{m_z / m_y}. \) Here we have used the polar coordinate \( k = (\hat{x} \cos \theta + \hat{y} \sin \theta) k. \) Noticeably, under a non-adiabatic driving, regardless of the anisotropy/isotropy of the Rashba SOC, the two components, \( \langle \sigma \rangle \parallel(k, t_1) \) and \( \langle \sigma \rangle \perp(k, t_1) \), of the driven spin texture, share similar characters in the field configuration over the angular part of \( k \) (see Eqs. (C1b) and (C1c), with the common factor of \( G_1 (\theta, \varphi_e), \) which is distinct from \( G_0 (\theta, \varphi_e) \) in the component \( \langle \sigma \rangle \parallel(k, t_1) \), Eq. (C1a). This correlation between the parallel-to-field in-plane component and out-of-plane component of the driven spin texture is not found in the adiabatic driven spin texture. Below we further investigate the consequences on the CISP effect from such driven spin textures.

a. Properties of the CISP orientation with isotropic Rashba SOC

Under the isotropic Rashba SOC, namely, \( r_A = 1 \), the driven spin texture is simplified to

\[
\langle \sigma \rangle \parallel(k, t_1) = 2 \delta s / \cos (2\theta - 2\varphi_e) - 1,
\]

\[
\langle \sigma \rangle \perp(k, t_1) = -2 \delta s / \sin (2\theta - 2\varphi_e),
\]
and
\[
\langle \sigma_z \rangle (\mathbf{k}, t_1) = -2\delta s_z \sin (2\theta - 2\varphi_e). \tag{C6c}
\]
By \( \int_0^\pi d\theta \sin (2\theta - 2\varphi_e) = \int_0^\pi d\theta \cos (2\theta - 2\varphi_e) = 0 \), we find \( S^\parallel (t_1) = S^z (t_1) = 0 \) and \( S^\perp (t_1) \neq 0 \). This result is summarised as
\[
S^z (t)|_{r_A=1} = 0, \tag{C7}
\]
\[
S (t) \cdot \hat{e}|_{r_A=1} = 0. \tag{C8}
\]

Here we should explain that the reason behind the vanishing of \( S^\parallel /z (t > t_0) \) under the isotropic condition, \( r_A = 1 \), is quite different from zero spin polarisation \( S_{eq} = S (t \leq t_0) = 0 \) given by the equilibrium spin texture regardless of the value of \( r_A \). The latter is simply due to \( \langle \sigma \rangle (\mathbf{k}, t \leq t_0) = 0 \) by the TRS while the driven spin texture generally satisfies \( \langle \sigma \rangle (\mathbf{k}, t > t_0) \neq 0 \). Noticeably from Eqs. (C6b) and (C6c), we find
\[
\langle \sigma_{\parallel /z} \rangle (\mathbf{k}, t_1) |_{\theta=\varphi_e+\delta\theta} = -\langle \sigma_{\parallel /z} \rangle (\mathbf{k}, t_1) |_{\theta=\varphi_e-\delta\theta}
\]
in which \( \varphi_e \) specifies the direction of the unit vector \( \hat{r}_S \) given by \( \hat{r}_S \parallel \mathbf{e} \) or \( \hat{r}_S \parallel R_{\pi/2} \hat{e} \) where \( R_{\pi/2} \) is a rotation by \( \pi/2 \) and \( \delta\theta \) is the angle between \( \mathbf{k} \) and \( \hat{r}_S \). So the isotropy of the Rashba SOC forces the driven spin texture in the \( \parallel \) and \( z \) components to become anti-symmetric in \( \theta \) with respect to \( \theta = \varphi_e \) causing \( S^\parallel (t_1) = S^z (t_1) = 0 \). Quite differently, we see from Eq. (C6a) that \( \langle \sigma_{\perp} \rangle (\mathbf{k}, t_1) \) becomes larger when \( \theta \) gets away from \( \varphi_e \) and it is maximised at \( \theta = \varphi_e \pm \pi/2 \).

The above points manifest that the external field along \( \hat{e} \) creates an anisotropic feature in the driven spin texture despite the Rashba SOC is isotropic. By numerically calculating the components of the driven spin texture under the same electric-field pulse used in the main text, we confirm that the above conclusion is held also to any later time \( t > t_0 \). The anti-symmetric character of the components \( \langle \sigma_{\parallel /z} \rangle (\mathbf{k}, t) \) and \( \langle \sigma_{\parallel /z} \rangle (\mathbf{k}, t) \) of driven spin texture with respect to the driving field orientation \( \hat{e} \) is demonstrated in Fig. 4(a) and Fig. 4(b) respectively. The dynamics of \( \langle \sigma_{\perp} \rangle (\mathbf{k}, t) \) is exemplified in Fig. 4(c). As expected from isotropic condition, we verified from the numerical results that the non-vanishing magnitude of \( S^\perp (t) \) is independent of the direction of \( \hat{e} \). Therefore, under the isotropic condition, the orientational properties, Eqs. (C7) and (C8) of CISP, caused by the non-adiabatic driving is not different from those arising from adiabatic drivings, namely, Eqs. (B10) and (B11). Our findings here are consistent with Ref. [4] in which a time-independent electric field is applied to study the time development of the spin polarisation in isotropic Rashba systems in the intrinsic limit.

Figure 4: Dynamics of driven spin texture components for selected \( \mathbf{k} \)'s. Writing in the polar coordinate \( \mathbf{k} = (k, \theta) \), we fix \( k = k_{\omega m} \) (see captions in Fig. 3 of the main text). The applied electric field is polarised at \( \varphi_e = 0.25\pi \). The directions of selected \( \mathbf{k} \)'s relative to the polarisation direction \( \hat{e} \) are illustrated on the upper-left corner of each plot. In (a) and (b), the anti-symmetry in \( \theta \) with respect to \( \varphi_e \) for the \( z \)- and \( \parallel \)-components are exemplified by choosing \( \theta = \varphi_e + \delta\theta \) (blue dashed) and \( \theta = \varphi_e - \delta\theta \) (red solid) where we set \( \delta\theta = 0.05\pi \) and \( \varphi_e = \varphi_e \) for (a) and \( \varphi_e = \varphi_e + \pi/2 \) for (b). In (c), \( \theta \)'s are given by \( \theta = \varphi_e \) (black dotted) and \( \theta = \varphi_e + \delta\theta \) with \( \delta\theta = 0.15\pi \) for the green dashed, \( \delta\theta = 0.5\pi \) for the red solid and \( \delta\theta = 0.7\pi \) for the blue dash-dot lines. The same pulse as that used in Fig. 3 of the main text is applied here.
By lifting up the isotropy and restoring the anisotropy in the Rashba SOC which establishes the mirror symmetry as its native structure, one finds that the non-adiabatic drivings can yield results qualitatively different from the adiabatic drivings. The anisotropic Rashba SOC featured by $r_A \neq 1$ yields $S^\parallel (t) \propto \sin (2\phi_e)$ [31], which is nonzero when $\hat{e}$ is not parallel to either of two the mirror lines $\hat{x}$ and $\hat{y}$. In other words, by allowing anisotropy in Rashba SOC so the system’s symmetry is featured by the two mirrors, a non-adiabatic driving field can be oriented to break this mirror symmetry and induces nonzero spin polarisation in both $S^\parallel (t)$ and $S^\perp (t)$ components. In contrast, in the adiabatic regime, even when the electric field is not applied along the mirror lines, it cannot induce an out-of-plane spin polarisation $\hat{z} \cdot S^{ad} (t) = 0$. We have demonstrated the induction of the out-of-plane CISP by transient mirror-symmetry breaking without excluding extrinsic scatterings in Fig. 3(a) of the main text. Here, we illustrate this intrinsic mechanism in the intrinsic limit in Fig. 5(a) and (b). It testifies that the more the external field’s polarisation is away from the mirror lines, the higher the peak values of $S^\parallel (t)$ can be attained (see the larger magnitudes in the red dashed lines relative to other lines there). The CISP component $S^\perp (t)$ which is not induced due to breaking of the mirror symmetry by the applied field is supplemented in Fig. 5(c). The black solid and blue dash-dot lines in all plots of Fig. 5 are obtained by deviating $\hat{e}$ from $\hat{x}$ and $\hat{y}$ by the same angle. Their difference attests the anisotropy between $\hat{x}$- and $\hat{y}$- directions. This anisotropic effect is then present in all three spin polarisation components as shown in Fig. 5.

![Figure 5: Intrinsic CISP dynamics for anisotropic Rashba SOC. The demonstrated polarisation angles $\varphi_e$’s and all other parameters are the same as those used in Fig. 3 of the main text except here all scattering rates are zero. The directions of applied field’s polarisation $\hat{e}$ with respect to the mirror lines $\hat{x}$ and $\hat{y}$ are illustrated as the inset of (a). When $\hat{e}$ lies in the middle between $\hat{x}$ and $\hat{y}$ corresponding to $\varphi_e = 0.25\pi$ (the red rod in the inset), the applied field maximally breaks the mirror symmetry and thus gives the highest peak values in $S_\perp (t)$ and $S_\parallel (t)$ but not in $S^\parallel (t)$ (see the red dashed lines in comparison to the other two lines in all three plots).](image)

2. Extraction of spin-mediated processes in the photocurrents by mirror-symmetry breaking

Having discussed the pulse-induced dynamics of the spin polarisation, here we provide accompanied studies of the photocurrent. In the main text, we have suggested that the spin-mediated part of the photocurrent may contain SOC-geometric effects in Sec. IIIC3 without specifying the SOC type. Here we discuss such manifestation with the Rashba SOC explicated in Sec. IIIB of the main text.

Substituting Eqs. (33) and (34) into Eqs. (32b) and (32c) from the main text, we obtain

$$j_K (t) = -e\chi_K \int a^2 k^2 \hbar \hat{k}_i \frac{\hbar k}{m_r} N_k (t),$$

$$j_{so} (t) = \left( -\frac{e}{\hbar} \right) \frac{\hbar k_{so}}{m_r} \hat{z} \times \chi_S S (t),$$

where $\chi_S = \begin{pmatrix} r_Y & 0 \\ 0 & r_X \end{pmatrix}$, $m_r = \sqrt{m_x m_y}$, $N_k (t) = \text{tr} (\varphi_k (t))$ and we have defined $\hbar k_{so} / m_r = \alpha_R / \hbar$ with $\alpha_R$ being the Rashba coefficient in Eq. (34) of the main text. The SOC-geometric effects are thus carried by $j_{so} (t)$ when the
spin polarisation, $\mathbf{S}(t) \neq 0$, can be attributed to the intrinsic spin coherence induced by the external field. Previous discussion already shows that using the asymmetry offered by $r_A \neq 1, \hat{e} || \hat{x}/\hat{y}$, one can tell, by the qualitative difference between $S^z(t) = 0$ and $S^z(t) \neq 0$ (the out-of-plane macroscopic spin polarisation), whether or not the macroscopic current $j_{so}(t)$ (which is coupled to the in-plane spin polarisation $S^{\parallel/\perp}(t)$) contains intrinsic spin coherence effects. We now investigate the implications of $r_A \neq 1, \hat{e} || \hat{x}/\hat{y}$ for the extraction of spin-mediated processes in the transient photocurrent $j(t, \varphi_e) = \hat{e}j^\parallel(t, \varphi_e) + (\hat{z} \times \hat{e}) j^\perp(t, \varphi_e)$ in which $j^\parallel(t, \varphi_e)$ and $j^\perp(t, \varphi_e)$ are called the longitudinal and the transverse components respectively. We make the dependence of the photocurrent on laser polarisation $\varphi_e$ explicit.

a. Transverse component as a mirror-symmetry-breaking signature

Explicitly, the the longitudinal and the transverse components of $j_K(t)$ from Eq. (C9) read

$$j^\parallel_K(t, \varphi_e) = -\epsilon \frac{\hbar K_i}{m_r} \tilde{n}_f \left[ \tilde{r} - \delta r \cos (2\varphi_e) \right],$$  \hspace{1cm} (C11a)

$$j^\perp_K(t, \varphi_e) = -\epsilon \tilde{n}_f \frac{\hbar K_i}{m_r} \delta r \sin (2\varphi_e),$$  \hspace{1cm} (C11b)

in ignorance of scattering effects. $\varrho_k(t) \rightarrow \varrho_k^{eq}(t)$. Here $\tilde{n}_f = \int d^2 \mathbf{k} \varrho_k^{eq}$ is the carrier density. Meanwhile, the components of $j_{so}(t)$ from Eq. (C10) can be written as

$$j^\parallel_{so}(t, \varphi_e) = \left( -\epsilon \frac{\hbar k_{so}^+}{\hbar^2/2} \right) \delta r \sin (2\varphi_e) S^\parallel(t, \varphi_e) + \cos (2\varphi_e) S^\perp(t, \varphi_e)$$

$$j^\perp_{so}(t, \varphi_e) = \left( -\epsilon \frac{\hbar k_{so}^+}{\hbar^2/2} \right) \delta r \cos (2\varphi_e) S^\parallel(t, \varphi_e) + \cos (2\varphi_e) S^\perp(t, \varphi_e).$$  \hspace{1cm} (C12a)

Clearly, by setting $\delta r = 0$ (so $r_A = 1$) in Eqs. (C11) and (C12), the transverse components vanish $j^\perp_K(so)(t, \varphi_e) = 0$ (recalling that $S^\parallel(t, \varphi_e)|_{r_A = 1} = 0$) and only the longitudinal components remain. When $\delta r \neq 0$ and at the same time $\sin (2\varphi_e) \neq 0$, which is equivalent to $r_A \neq 1, \hat{e} || \hat{x}/\hat{y}$, then the possibility $j^\perp_K(so)(t, \varphi_e) \neq 0$ arises. The transverse component $j^\perp(t, \varphi_e)$ thus is a signature of breaking the mirror symmetry. Note that $j^\parallel_K(so)(t, \varphi_e)$ is invariant under the mirror reflection operations $\varphi_e \rightarrow -\varphi_e$ and $\varphi_e \rightarrow \pi - \varphi_e$.

b. Differentiating spin- and bond-mediated processes via the longitudinal and transverse photocurrents with anisotropy

The mirror symmetry here is associated with the anisotropy in the Rashba SOC by $r_A \neq 1$. The efficacy of the anisotropy in principle can be quantified by the difference between the results obtained under $\hat{e} || \hat{x}$ and that obtained under $\hat{e} || \hat{y}$. However, since the transverse components of the current need to be induced by deviating $\hat{e}$ from $\hat{x}$ and $\hat{y}$, we instead measure the efficacy of the anisotropy by the difference between $\hat{e} || R_z(\delta \varphi) \hat{x}$ and $\hat{e} || R_z(-\delta \varphi) \hat{y}$. Here $R_z(\delta \varphi) \hat{x}$ rotates $\hat{x}$ by an angle $\delta \varphi$ with $0 < \delta \varphi \leq \pi/4$ toward $\hat{y}$ (so $\varphi_e = \delta \varphi$) and correspondingly $R_z(-\delta \varphi) \hat{y}$ rotates $\hat{y}$ by the same amount of $\delta \varphi$ toward $\hat{x}$ ($\varphi_e = \pi/2 - \delta \varphi$). This leads to the definition of anisotropic asymmetry as Eq. (36) in the main text. We now study the anisotropic asymmetry in the longitudinal ($\alpha = ||$) and transverse ($\alpha = \perp$) components of the bond- and spin-mediated parts of the photocurrents given by Eqs. (C11) and (C12) one by one.

To simplify the analysis of the anisotropic asymmetry, we considered $\delta \varphi \gtrsim 0$ such that $\sin (2\delta \varphi) \approx 0$ and $\cos (2\delta \varphi) \approx 1$. Then, applying Eq. (36) to Eqs. (C11) and (C12) and taking $\alpha = ||$, we obtain

$$\Delta \delta \varphi j^\parallel_K(t) \approx \epsilon \frac{\hbar K_i}{m_r} \left( r_Y - r_X \right) \tilde{n}_f,$$  \hspace{1cm} (C13a)

$$\Delta \delta \varphi j^\parallel_{so}(t) \approx \left( -\epsilon \frac{\hbar k_{so}^+}{m_r} \right) \left( \frac{1}{\hbar^2/2} \right) \left[ r_Y S^\perp(t, \pi/2 - \delta \varphi) - r_X S^\perp(t, \delta \varphi) \right].$$  \hspace{1cm} (C13b)
Note here that Eq. (C13a) is an exact consequence of Eq. (C11b) without any use of approximations. The approximation \( \delta \phi \geq 0 \) has been applied to simplify Eq. (C12b) to arrive Eq. (C15b). In general for \( \delta \phi \neq 0 \) and \( \delta r \neq 0 \) we have non-vanishing \( \Delta_{\delta \phi j^+_K} (t) \neq 0 \). The sharp contrast between the spin-mediated part \( \Delta_{\delta \phi j^+_K} (t) \neq 0 \) and the bond-mediated part \( \Delta_{\delta \phi j^+_K} (t) = 0 \), and therefore \( |\Delta_{\delta \phi j^+_K} (t) = \Delta_{\delta \phi j^+_K} (t) | \gg |\Delta_{\delta \phi j^+_K} (t) | = 0 \), here obtained under the intrinsic limit, is directly linked to the difference between the \( V^K (k_t) \) and \( V^{so} (k_t) \) in the definition of the velocity matrix Eq. (31) in the main text. The inclusion of scattering effects does not affect the contents of \( V^K (k_t) \) and \( V^{so} (k_t) \) at all. It only modifies the dynamics of \( g_k (t) \) so one could still have

\[
|\Delta_{\delta \phi j^+_K} (t) = \Delta_{\delta \phi j^+_K} (t) | \gg |\Delta_{\delta \phi j^+_K} (t) | ,
\]

with \( \Delta_{\delta \phi j^+_K} (t) \) being not strictly zero, as verified from numerical calculations shown in Fig. 3(c) of the main text.

Combining Eqs. (C16) and (C14), we see that the spin/bond-mediated part of the photocurrent is then largely contained in its transverse/longitudinal component of anisotropic asymmetry. Noticeably, \( K_t \gg k^*_{so} \) for holding (C14) is another indication of the activation of the non-adiabatic regime of spin dynamics. So the distinction between bond-mediated and the spin-mediated parts of the photocurrent manifested by the distinction between the longitudinal and the transverse components of the photocurrents, in terms of the anisotropic characters, is crucially facilitated by non-adiabatic drivings.

**Appendix D: Supporting information for implications on experiments**

Implications of NADT-WPT to experiments with ultrafast lasers applied to quantum materials have been discussed in Sec. IVB of the main text in two parts. In the first part, we point out the general existence of links between the present theoretical framework to three kinds of experiments without going to particular points specific to each type of experiments. These particular points are discussed in more details for each of the three types of experimental detections in Sec. VIB here. In the second part of Sec. IVB of the main text, we summarised theoretical implications relevant for exploiting SOC anisotropic effects in experiments. For this, we have performed THz-emission experiments on SnSe. Therefore, in Sec. VII here, we describe experimental details and how the consistency between theoretical implications and experimental observations is reached.

1. **General considerations of relating NADT-WPT to experiments with ultrafast lasers**

   a. **Time-resolved ARPES (Tr-ARPES)**

   The first type concerns the Tr-ARPES. With subcycle time resolution [44], the Tr-ARPES is capable of tackling what we call the in-pulse time region here. From the time-dependent data of occupation distributed over energy and
momentum, the group velocities of the bands and the dynamics of Bloch-momentum-resolved intra-band processes can be tracked. These capabilities of Tr-ARPES have been demonstrated to manifest the intrinsic properties of the linearly dispersed surface Dirac cones in experiments [14]. Here we seek for temporal dynamics of momentum-resolved inter-band coherence rooted to inter-band geometric connections. The challenge, therefore lies in getting the transient inter-band coherence and not just the occupation. With the advancement of spin-resolved ARPES [92, 93], it may be possible to extract information about spin coherence from the spin-resolved-ARPES data to microscopically address the SOC-geometric effects in real time.

b. Time-averaged electronic currents

The second type is implemented by attaching electrodes to measure the laser-excited currents. Given the limitation on the response time of the electronic devices, the time-averaged currents, integrated on a time scale larger than the microscopic time scale of the excited electron dynamics, are often obtained. The information contained in such currents is further complicated by laser-spot-to-collector diffusions as well as the interfaces between the sample quantum material and the electrodes. Nevertheless, through a number of studies on graphene under strong-field drivings where the non-adiabatic dynamics is relevant, an important experimental observation has been confirmed. That is, the time-averaged currents carry the imprints of the subcycle properties of the driving fields [94 - 96]. In understanding these results, the current is formulated in terms of the asymptotic conduction population and the group velocity [97]. The development of NADT-WPT made here, without prior references to the above works, is unexpectedly found to be related to the above results when we tried to look for implications of NADT-WPT on experiments. We analytically showed that the post-pulse current has a history dependence on the in-pulse non-adiabatic dynamics arising from the geometric connections among the bands. The final result is akin to the dependence of post-pulse time-averaged currents on the parameters of the in-pulse drivings observed in experiments which can be literally interpreted also as history-dependence effects. The current, from a wave-packet point of view, however, is not only contributed by the group velocities of bands but also by the anomalous velocities supported by inter-band coherence. To which extent these two different sources of currents survive the time-averaging processes and carry history information of in-pulse-drivings despite of spot-to-contact diffusion and other complications remain to be investigated in the future.

c. Time-resolved THz emissions

The third type measures the THz-emission associated with the laser-excited photocurrents. In principle, one considers transient currents in this case, instead of time-integrated currents. Within the framework of electromagnetism without any explicit band structure consideration, the electric fields of the emitted signals $E_{em}(t)$ have been shown to be proportional to $dj(t)/dt$ [82]. This seems to directly relate experimental signals to the present theory which computes $j(t)$ and $dj(t)/dt$. Nevertheless, excitations other than the sought-for geometrically-rooted intrinsic inter-band coherence can be induced. Detected transient signals in the time domain can be time-delayed from the photoexcitations due to inelastic relaxations. The resulting emission then contains more information than just $dj(t)/dt$ produced from the theory (even when it includes finite scattering rates) used here. Therefore with these caveats in mind, to relate NADT-WPT to THz emissions, one shall not directly compare the detailed time dependent profiles of experimentally produced $E_{em}(t) \sim dj(t)/dt$ with that of a clean theory result. Instead, it is more suitable to relate the transient dynamics of currents given by NADT-WPT to THz emissions. This gives a more suitable way to relate views of NADT-WPT to THz emissions data as we demonstrate below.

2. THz-emission experiments on SnSe viewed from NADT-WPT

We have collected THz-emission data as a result of applying ultrafast laser pulses on single crystal SnSe. The experimental methods are described in Sec. D2a. Analysis of the experimental data in line with implications from NADT-WPT summarised in Sec. IV of the main text is conducted in Secs. D2b and D2c. There, we have employed
a. Sample preparation and experimental methods

The SnSe single crystals were prepared using the Bridgman method. Tin rods (99.99%) and Selenium beads (99.999%) were used as starting materials. Stoichiometric amounts of raw elements were loaded into a small quartz tube and sealed into a second ampoule to avoid cracking upon cooling. The charged ampoule was heated in a vertical furnace to 1183 K in 12 hr. and kept at 1183 K for 24-48 hr. for homogenisation. After that, the ampoule was slowly cooled to 973 K at a rate of 1 K/h and kept for 50 hr. Millimetre-sized crystals with metallic luster were obtained. The crystal structure was confirmed by X-ray diffraction (XRD) with Cu Kα radiation.

A Ti:sapphire regenerative amplifier (Astrella UPS, Coherent, Inc.) was employed to perform the THz emission experiments. An optical pulse train with a central wavelength of 800 nm (∼1.55eV), a pulse duration of ∼35 fs, and a repetition rate of 1 kHz was split into a pump beam and a probe beam. The pump beam (average power ∼ 1.5 mW) was used to illuminate the surface of SnSe single crystals to generate THz electromagnetic radiation. The probe beam was used in electro-optic sampling to depict the THz waveform in the time domain by changing the optical path difference between the pump and probe beams. A wire-grid polariser was used to purify the polarisation of the emitted THz radiation from samples in vertical (S-wave) or horizontal directions (P-wave). See detection layouts in Fig. 6(a), where the unit vectors along the three crystal axes, namely, a-, b- and c-axes are denoted by \( \hat{a} \), \( \hat{b} \) and \( \hat{c} \) respectively. Corresponding atomic structure of the single crystal SnSe is schematically shown in Fig. 6(b).

We apply laser pulses linearly polarised (LP) in the lab-referenced fixed direction \( \hat{\ell} \), perpendicular to the incident plane (shallow blue) spanned by the incoming and outgoing propagation vectors (the orange and the deep-blue solid arrows respectively) of the electromagnetic waves (see Fig. 6(a)). The incident electric fields of the LP pulses parallel to \( \hat{\ell} \) thus lie on the a-b plane of the sample (coloured in yellow, see Fig. 6(a)). The plane spanned by the a- and b- crystal axes is called the sample plane throughout this work. The angle \( \varphi_{in} \) between the crystal a-axis and the fixed direction \( \hat{\ell} \) can be varied by rotating the sample plane around the c-axis. The electric fields of the emitted S-wave are along the direction \( \hat{\ell} \) (see the double-headed red arrow in Fig. 6(a)). The electric fields of the emitted P-wave (see the double-headed brown arrow in Fig. 6(a)) are perpendicular to \( \hat{\ell} \). The LP-pulse induced S-wave and P-wave with the sample rotation angle \( \varphi_{in} \) are denoted respectively by \( E^s_{em}(t, \varphi_{in}) \) and \( E^p_{em}(t, \varphi_{in}) \). We also apply right-/left-hand circularly polarised (CP) pulses \( \sigma_+ / \sigma_- \). The corresponding THz emissions as S-wave and P-wave are denoted by \( E^s_{em}(t, \varphi_{in}) \) and \( E^p_{em}(t, \varphi_{in}) \), respectively and the circular-dichroism (CD) signals are defined by

\[
E^s_{CD}(t) := E^s_{em}(t, \sigma_+) - E^s_{em}(t, \sigma_-)
\]

\[
E^p_{CD}(t) := E^p_{em}(t, \varphi_{in})
\]

b. The a-b plane of SnSe single crystals as the playground for the in-plane SOC

We first confirm that the plane on which the SOC plays a major role is the a-b plane of SnSe single crystals. Two different methods have been used to reach the same conclusion that the c-axis signals are not important with regards to our interests here. The first method uses CD data only. The second method uses data from LP excitations only with auxiliary analysis guided by NADT-WPT.

The S-wave CD data \( E^s_{CD}(t) \) obtained with \( \varphi_{in} = 0 \) and \( \varphi_{in} = \pi/2 \) represent the SOC effects encoded in the emitted waves along the a-axis and b-axis respectively. They are shown respectively as the black solid (for a-axis) and the red dashed (for b-axis) lines in Fig. 6(c). In accompanying to this, the P-wave CD data \( E^p_{CD}(t) \) obtained with \( \varphi_{in} = 0 \) and \( \varphi_{in} = \pi/2 \) show the CD emission waves on the b-c plane and the a-c plane, respectively plotted as the red dashed and the black solid lines in Fig. 6(d). By comparing the overall magnitudes of the a-axis signal in Fig. 6(c) with that of the a-c plane signal in Fig. 6(d) (both as black solid lines), we find the involvement of the c-axis signal does little to affect the observed CD magnitudes contributed by the signal fields along the a-axis. Similar conclusions can be drawn from the comparison between the b-axis signal in Fig. 6(c) with that of the b-c plane in Fig. 6(d) (both as red dashed lines). Therefore, the SOC-related electronic motion is mainly on the a-b plane, which is also called the sample plane here. We extract the peak-to-peak values from the time sequences of \( E^s_{CD}(t) \) obtained for the a-axis and b-axis respectively denoted as \( E^s_{CD} \). They exhibit obvious anisotropy, namely, \( E^s_{CD} \approx 1 \mu V > E^p_{CD} \approx 0.2 \mu V \).

The time-resolved THz emissions induced by LP pulses applied on the sample plane with the sample rotation angle \( \varphi_{in} \) scanned over a round are documented in Fig. 6(e). From these data we deduce the in-plane (on the sample-plane) longitudinal and transverse components of the emitted THz electric fields in line with the NADT-WPT treatment of the photocurrent induced by LP pulses in terms of its spatial components || and ⊥ to \( \hat{e} \) in the main text. The in-plane anisotropy here is furnished by two orthogonal and non-equivalent axes, namely, the a- and b-axes, showing similarity particular data extraction procedures to separate away the signal components relevant to our investigation purpose from those uninterested components. These fitting procedures guided by NADT-WPT were detailed in Sec. 33.
to Sec. III B We henceforth tried Eq. (38) of the main text to construct mirror-symmetric and mirror-asymmetric functions as fitting functions for the emitted in-plane electric fields that vary longitudinally and transversely to \( \hat{\ell} \) respectively. Details are in Sec. I D 2 d. The longitudinal components are given by the electric fields of the S-wave \( E_{em}^x (t, \varphi_{in}) \) since they extend in parallel to \( \hat{\ell} \). The suitability of mirror-symmetric fitting \( E_{cm}^y (t, \varphi_{in}) \) for \( E_{em}^y (t, \varphi_{in}) \) is demonstrated in Fig. 7(a). The directly detectable P-wave \( E_{cm}^p (t, \varphi_{in}) \) contains both an uninterested \( c \)-axis component (the double-headed green arrow in parallel with \( \hat{c} \) in Fig. 6(a)) and the targeted component \( E_{em}^z (t, \varphi_{in}) \) (the double-headed yellow arrow in Fig. 6(a)). A particular procedure (see Sec. I D 2 d) is thus applied to fix this \( c \)-axis component with direct fitting to \( E_{em}^z (t, \varphi_{in}) \) by either mirror-symmetric \( f_A^p (t, \varphi_{in}) \) or mirror-asymmetric \( f_A^p (t, \varphi_{in}) \) functions. From the above procedure, then \( E_{em}^z (t, \varphi_{in}) \) is extracted. An improvement using the mirror-asymmetric function \( f_A^p (t, \varphi_{in}) \) over the mirror-symmetric function \( f_S^p (t, \varphi_{in}) \) to fit \( E_{em}^p (t, \varphi_{in}) \) is demonstrated in Fig. 7(b). The mirror-symmetric/asymmetric contrast \( E_{cm}^y (t, \varphi_{in}) / E_{em}^y (t, \varphi_{in}) \) is showcased in Fig. 7(c) along with a reference of similar contrast shown in Fig. 7(d) from the theoretical calculation of the Rashba system of Sec. III B in the main text.

From the time sequences of \( E_{em}^z (t, \varphi_{in}) \), \( E_{em}^y (t, \varphi_{in}) \) and \( E_{em}^z (t, \varphi_{in}) \), we extract their peak-to-peak values at each angle \( \varphi_{in} \) and they are denoted by \( E_{cm}^z (\varphi_{in}) \), \( E_{em}^y (\varphi_{in}) \) and \( E_{em}^z (\varphi_{in}) \) respectively. These peak-to-peak values are documented in Fig. 7(e). The closeness of \( E_{cm}^z (\varphi_{in}) \) (green triangles) to \( E_{em}^z (\varphi_{in}) \) (black crosses) in Fig. 7(e) indicates that the \( c \)-axis components contained in \( E_{em}^z (\varphi_{in}) \) are relatively small. This conclusion about the insignificance of the \( c \)-axis component, obtained purely from the LP data aided by fittings in accordance with NADT-WPT, is in line with the similar conclusion that is purely obtained from the CD data above.
We are now facilitated with enough information to inspect the theoretical points (i-iii) abstracted in Sec. IVB of the main text. Theoretically the logic usually flows as (i)+(ii)=>(iii), started by the physical picture of (i) depicting the spin-mediation and the bond-mediation of electronic currents as fundamentally different processes. However, from the perspective of data availability in experiments where it is only the full signal (spin-mediated plus bond-mediated ones) that is accessed directly, it is operationally more convenient to first verify (ii) and (iii). Then by plain logic, (i) directly follows from (ii)+(iii). Below we discuss how the consistency between the points (ii-iii) and the experimental data is observed. For tackling (ii), we study the differentiation between longitudinally and transversely excited THz emissions under LP pulses, in terms of their respective anisotropic properties. This can be approached with or without the involvement of the CD data.

We first go without consulting the CD data by directly inspecting the $\varphi_{in}$-dependence patterns displayed in the transient processes and in the peak-to-peak extractions. For the longitudinal component, both the transient snapshots $E_{\parallel}^m(t, \varphi_{in})$ and the peak-to-peak values $E_{\parallel}^\parallel(\varphi_{in})$ exhibit visually identifiable mirror-symmetric pattern (see the red line and the red disks in Fig. 7(c) and (e) respectively). In contrast, for the transverse component, only the peak-to-peak extractions $E_{\perp}^\parallel(\varphi_{in})$ show a more mirror-symmetric pattern which is apparently not visible in the snapshots $E_{\perp}^m(t, \varphi_{in})$ (see the green triangles and the green line in Fig. 7(e)) respectively. This observation of the contrast between the longitudinally and transversely induced signals corroborates that longitudinal and transverse currents are excited by underlying different mechanisms, abstracted as the point (ii) for Sec. IV in the main text.

We now describe how the CD data can be useful. In parallel to the theoretical view of Sec. IIC3 of the main text, the emitted electric fields lying on the sample plane are also decomposed to two parts respectively attributed to the underlying bond-mediated (with subscript $K$) and the spin-mediated (with subscript $so$) processes behind the excited photocurrents, namely,

$$E_{\parallel,\perp}^\parallel (\hat{\ell}) = E_{\parallel,\perp}^{\parallel/K}(\hat{\ell}) + E_{\parallel,\perp}^{\parallel/so}(\hat{\ell}) .$$  

(D1)

In Eq. (D1), we replace $\varphi_{in}$ that appear in the arguments of the LP-pulse-induced signals by $\hat{\ell}$ for convenience of later discussions. The previously obtained anisotropic relation in CD signals $E_{\parallel,\perp}^{\parallel/K} > E_{\parallel,\perp}^{\parallel/so}$, is first used to deduce similar anisotropic relations for the spin-mediated parts of the LP-induced emissions according to the link between CD and SOC [97]. Then together with Eq. (D1), we can attempt to infer possible anisotropic relations for the bond-mediated parts to deduce further differentiation between the spin-mediated and the bond-mediated contributions in LP-induced THz emissions.

The CD detection is a common experimental mean to certify the existence of SOC [97] that also underlies the longitudinal and transversal THz emissions under LP pulses, in terms of their respective anisotropic properties. This can be approached with or without the involvement of the CD data. Theoretically the logic usually flows as (i)+(ii) => (iii), started by the physical picture of (i) depicting the spin-mediation and the bond-mediation of electronic currents as fundamentally different processes. However, from the perspective of data availability in experiments where it is only the full signal (spin-mediated plus bond-mediated ones) that is accessed directly, it is operationally more convenient to first verify (ii) and (iii). Then by plain logic, (i) directly follows from (ii)+(iii). Below we discuss how the consistency between the points (ii-iii) and the experimental data is observed. For tackling (ii), we study the differentiation between longitudinally and transversely excited THz emissions under LP pulses, in terms of their respective anisotropic properties. This can be approached with or without the involvement of the CD data.

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$$E_{\parallel,\perp}^\parallel (\hat{\ell}) = E_{\parallel,\perp}^{\parallel/K}(\hat{\ell}) + E_{\parallel,\perp}^{\parallel/so}(\hat{\ell}) .$$  

(D1)

In Eq. (D1), we replace $\varphi_{in}$ that appear in the arguments of the LP-pulse-induced signals by $\hat{\ell}$ for convenience of later discussions. The previously obtained anisotropic relation in CD signals $E_{\parallel,\perp}^{\parallel/K} > E_{\parallel,\perp}^{\parallel/so}$, is first used to deduce similar anisotropic relations for the spin-mediated parts of the LP-induced emissions according to the link between CD and SOC [97]. Then together with Eq. (D1), we can attempt to infer possible anisotropic relations for the bond-mediated parts to deduce further differentiation between the spin-mediated and the bond-mediated contributions in LP-induced THz emissions.

The CD detection is a common experimental mean to certify the existence of SOC [97] that also underlies the LP-induced signals $E_{\parallel,\perp}^{\parallel/K}(\hat{\ell})$. The result $E_{\parallel,\perp}^{\parallel/K} > E_{\parallel,\perp}^{\parallel/so}$ implies that the SOC effect is stronger for electronic motion along the direction of $\hat{a}$ than that in the direction of $\hat{b}$. Therefore, for longitudinally induced signals, we have

$$E_{\parallel/\perp}^{\parallel/so}(\hat{\ell} \parallel \hat{a}) > E_{\parallel/\perp}^{\parallel/so}(\hat{\ell} \parallel \hat{b}) .$$  

(D2)

For the transverse response, note that $E_{\parallel/\perp}^{\parallel/K}(\hat{\ell} \parallel \hat{a})$ is associated with currents that run in the direction of $\hat{b}$ and $E_{\parallel/\perp}^{\parallel/K}(\hat{\ell} \parallel \hat{b})$ is emitted by current that runs along $\hat{a}$. The implication from $E_{\parallel/\perp}^{\parallel/K} > E_{\parallel/\perp}^{\parallel/so}$ for transversely excited THz emissions by LP pulses should then be read as

$$E_{\parallel/\perp}^{\parallel/so}(\hat{\ell} \parallel \hat{b}) > E_{\parallel/\perp}^{\parallel/so}(\hat{\ell} \parallel \hat{a}) .$$  

(D3)

With Eqs. (D2) and (D3) for the spin-mediated parts and knowing also the full signals $E_{\parallel,\perp}^{\parallel/\perp}(\hat{\ell}) = E_{\parallel/\perp}^{\parallel/\perp}(\hat{\ell}) + E_{\parallel/\perp}^{\parallel/\perp}(\hat{\ell})$ in Fig. 7(e), we should be able to deduce corresponding relation between $E_{\parallel/\perp}^{\parallel/\perp}(\hat{\ell} \parallel \hat{a})$ and $E_{\parallel/\perp}^{\parallel/\perp}(\hat{\ell} \parallel \hat{b})$ as well as the relation between $E_{\parallel/\perp}^{\parallel/K}(\hat{\ell} \parallel \hat{a})$ and $E_{\parallel/\perp}^{\parallel/K}(\hat{\ell} \parallel \hat{b})$. For the longitudinal component, the full signals satisfy

$$E_{\parallel,\perp}^{\parallel/\parallel}(\hat{\ell} \parallel \hat{a}) < E_{\parallel,\perp}^{\parallel/\parallel}(\hat{\ell} \parallel \hat{b}) .$$  

(C4)

On the contrary, for the transverse components for which the full signals satisfy $E_{\parallel,\perp}^{\parallel/\parallel}(\hat{\ell} \parallel \hat{a}) = E_{\parallel,\perp}^{\parallel/\parallel}(\hat{\ell} \parallel \hat{a}) + E_{\parallel/\perp}^{\parallel/\perp}(\hat{\ell} \parallel \hat{b}) < E_{\parallel,\perp}^{\parallel/\parallel}(\hat{\ell} \parallel \hat{b}) = E_{\parallel,\perp}^{\parallel/\parallel}(\hat{\ell} \parallel \hat{b}) + E_{\parallel/\perp}^{\parallel/\perp}(\hat{\ell} \parallel \hat{b})$ (see green triangles in Fig. 7(e)) no definite relation between

(E)
the bond-mediated parts $E_{K}^{b} (\hat{\ell} \parallel \hat{b})$ and $E_{K}^{a} (\hat{\ell} \parallel \hat{a})$ similar to Eq. [D4] can be deduced from the spin-mediated parts Eq. [D3].

The above contrast between $\parallel$ and $\perp$ components of the THz emissions attributed to underlying photocurrents is briefly interpreted as the following. The information deduction from the spin-mediated signals to the bond-mediated signals works in one way for the longitudinal components while it works in other ways for the transverse components. This is another side evidence for the fundamental difference between longitudinal and transverse responses as point (ii). Since such difference between $\parallel$ and $\perp$ components is facilitated by the differentiation between spin-mediated and bond-mediated processes, it is equivalent to the point (iii), namely, these two different photocurrent components respectively encode the spin/bond-mediated processes in quite different ways. Hereby we find the observed data consistent with the points (ii) and (iii) briefed in Sec. [IVB] of the main text. The point (i) there is a logical consequence of (ii) and (iii).

![Figure 7](image)

Figure 7: Differentiation between $\parallel$ and $\perp$ components of THz emissions. (a): Detected S-wave $E_{em}^{s} (t, \varphi_{in})$ data points (black circle and red cross marks) and fitting to them by mirror-symmetric functions $E_{em}^{0} (t, \varphi_{in})$ (black solid and red dashed lines) (see legends for selected values of $\varphi_{in}$). (b): Fitting to data of $E_{em}^{p} (t, \varphi_{in})$ (black cross marks) by mirror-symmetric function $f_{p}^{1} (t, \varphi_{in})$ (red dashed line) is improved by mirror-asymmetric function $f_{p}^{1} (t, \varphi_{in})$ (blue dotted line) illustrated with $\varphi_{in} = 0.9\pi$. (c): A mirror-symmetric/asymmetric contrast between $E_{em}^{0} (t, \varphi_{in})$ (the red line) and $E_{em}^{0} (t, \varphi_{in})$ (the green line) from fittings to the experimental data is snapshotted at $t = 3.1\mu s$ as a polar plot over varied $\varphi_{in}$. (d): Similar mirror-symmetric/asymmetric contrast is seen in the polar plot of $\varphi_{in}$-dependencies of $j^{1} (t, \varphi_{e})$ (the red line) and $j^{1} (t, \varphi_{e})$ (the green line) computed theoretically using the Hamiltonian in Sec. [IIIB] of the main text. This is snapshotted at $t - t_{0} = 2.4t^{*}$ and with the same parameters used for the calculations of Fig. [3] in the main text. In both (c) and (d), the zero and maximum radial lengths correspond to $-0.8$ and $1.0$ in their respective units $0.1mV$ for (c) and $(-e/\hbar)k_{s}^{0} A_{s}^{0}$ for (d). (e): Peak-to-peak THz emitted signals as a function of $\varphi_{in}$. The red disks, black crosses and green triangles mark respectively $\dot{E}^{0} (\varphi_{in}), \dot{E}^{p} (\varphi_{in})$ and $\dot{E}^{\perp} (\varphi_{in})$.

d. Analysis of anisotropic characters under the excitation of LP pulses

Here we analyse the characters of the anisotropic dependence of THz signals $E_{em}^{s} (t, \varphi_{in})$ and $E_{em}^{p} (t, \varphi_{in})$ on the linear polarisation angle $\varphi_{in}$ or simply denoted as $\varphi$. The reference axis from which $\varphi$ is measured is re-named as the $x$-axis. The axis orthogonal to the $x$-axis is of course called the $y$-axis as usual. Taking these two axes as the mirror lines, we first note from the angular scans of $E_{em}^{s/p} (t, \varphi)$ that the mirror symmetric character in $E_{em}^{s} (t, \varphi)$ is more visible than that in $E_{em}^{p} (t, \varphi)$ (see Fig. [3]e)). More explicitly, a mirror-symmetric function $f_{S} (t, \varphi)$ is defined by satisfying

$$f_{S} (t, -\varphi) = f_{S} (t, \varphi), f_{S} (t, \pi - \varphi) = f_{S} (t, \varphi),$$

in which the first/second of Eq. [D5] describes reflection about the $x/y$-axis. According to Sec. [IIIB] of the main text, the analytical form of the bond-mediated part of longitudinal current Eq. [C11a], which does not involve any
form of SOC, is simply a result of generic quadratic dispersion. Its dependence on $\varphi$ given by $\cos(2\varphi)$ satisfies the mirror-symmetric property of Eq. (D5). In order to have the fitting parameters to be optimised, then according to $\cos(2\varphi) = \cos^2\varphi - \sin^2\varphi$ we assume

$$f_S(t, \varphi) = \alpha_0(t) \cos^2\varphi + \alpha_1(t) \sin^2\varphi,$$

in which $\alpha_0(t)$ and $\alpha_1(t)$ are treated as the fitting parameters. For mirror-asymmetric functions $f_A(t, \varphi)$, the transverse components of Eqs. (C11) and (C12) suggest the use of $\cos(2\varphi)$ and $\sin(2\varphi)$ in the composition of $f_A(t, \varphi)$. Real physical observable sufficiently repeats itself when $\varphi$ is varied over $2\pi$. This suggests one includes the most basic periodic functions $\cos(\varphi)$ and $\sin(\varphi)$ for fittings to $E_{em}^s(t, \varphi)$. So two forms of $f_A(t, \varphi)$ are compared, namely,

$$f_{A1}(t, \varphi) = \beta_0(t) \cos\varphi + \beta_1(t) \sin\varphi,$$

for which only the $2\pi$-periodicity is considered and

$$f_{A2}(t, \varphi) = \beta_0(t) \cos\varphi + \beta_1(t) \sin\varphi + c_0(t) \cos(2\varphi) + c_1(t) \sin(2\varphi),$$

for which the particular mirror-asymmetric characters from the theoretical consideration in Sec. IIIIB of the main text are included too.

Here we clarify which fitting function is applied to fit which signal. By re-labeling $\alpha_{0/1}(t)$ in Eq. (D6) as $\alpha_{0/1}^a(t)$ and renotating $f_S(t, \varphi)$ by $f_S^a(t, \varphi)$ as specification to the S-wave, the data $E_{em}^a(t, \varphi)$ is fitted by $f_S^a(t, \varphi)$. For the purpose of comparison, $E_{em}^p(t, \varphi)$ is fitted by three different functions, $f_S^p(t, \varphi), f_{A1}^p(t, \varphi)$ and $f_{A2}^p(t, \varphi)$ whose forms are given respectively by Eqs. (D6), (D7) and (D8) with the fitting parameters specified for the P-wave data. Note that the plane of interest is the $x$-$y$ plane but the P-wave contains also an out-of-plane $z$-component as mentioned before (recall the $c$-axis signal from Fig. 6(a)). By the theory of electromagnetism in medium considering the $x$-$z$ plane to be incident plane, we have [82],

$$E_{em}^p \propto \vec{J}_z - \gamma \vec{J}_x,$$

(D9)

where the current $\vec{J}$ has all three $x$-, $y$- and $z$-components. Here $\vec{J}_x$ and $\vec{J}_z$ are time derivatives of the $x$- and the $z$-components of the current. The coefficient $\gamma$ is related to the dielectric constant of the material [82]. Because $\varphi$ is an attribute of vectors on the $x$-$y$ plane orthogonal to the $z$-axis, we assume that $J_z$ is independent of $\varphi$. So fittings to $E_{em}^p(t, \varphi)$ are attempted by $f_{A1}^p(t, \varphi) = f_{A1}(t, \varphi) + f_1(t)$ for $i = 1, 2$ where $f_1(t)$ having no dependence on $\varphi$ accounts for $J_z$. The fitting parameters are found by minimising the difference between the values obtained by the fitting functions and those of the data themselves. The difference functions are thus defined as $L^s(t) = \sum_{n=1}^{N_{em}} \left| f_S(t, \varphi_n) - E_{em}(t, \varphi_n) \right|^2$ for the S-wave data and $L^p(t) = \sum_{n=1}^{N_{em}} \left| f_X(t, \varphi_n) - E_{em}(t, \varphi_n) \right|^2$ for the P-wave data, where $N_{in}$ is the number of angle values collected. These difference functions are non-negative. Here $X = \{S, A, 1, 2\}$ enumerates over different settings of $\varphi$-dependence characters for fitting the P-wave data. The fitting parameters $\{\alpha_{0/1}^a(t), \alpha_{0/1}^s(t)\}$ in $f_S^a(t, \varphi)$ are obtained by solving the algebraic equations $\partial L^s(t)/\partial \alpha_{0/1}^a(t) = 0$ and $\partial L^s(t)/\partial \alpha_{0/1}^s(t) = 0$ for given $t$. Similar procedure is applied to obtain the fitting parameters in $f_X^p(t, \varphi)$. To calibrate the error of fitting as dimensionless quantities, we define $\Delta E_{n/p} = \max \left\{ E_{em}^{n/p}(t_i, \varphi_n) \right\}_{i=1}^{N_T} - \min \left\{ E_{em}^{n/p}(t_i, \varphi_n) \right\}_{i=1}^{N_T}$ where $N_T$ is the number of points in the time sequence probed for the experimental run with fixed polarisation angle $\varphi_n$. The error at each tested angle $\varphi_n$ is then given by

$$\delta_n^s = \left[ \sum_{i=1}^{N_T} \left| f_S^a(t_i, \varphi_n) - E_{em}^a(t_i, \varphi_n) \right|^2 / N_T \right] / \Delta E_{n}^s$$

for fitting to the S-wave and $\delta_n^{pX} = \left[ \sum_{i=1}^{N_T} \left| f_X^p(t_i, \varphi_n) - E_{em}^p(t_i, \varphi_n) \right|^2 / N_T \right] / \Delta E_{n}^p$ for fitting to the P-wave. The overall error over all angles $\{\varphi_n\}_{n=1}^{N_{in}}$ is given by the averages over these non-negative values, namely, $e_n = \sum_{n=1}^{N_{in}} \delta_n^s / N_{in}$ and $e_{pX} = \sum_{n=1}^{N_{in}} \delta_n^{pX} / N_{in}$. To see if the error is particularly large for some values of $\varphi_n$, standard deviations in the error distributions $\{\delta_n^s\}_{n=1}^{N_{in}}$ and $\{\delta_n^{pX}\}_{n=1}^{N_{in}}$ are extracted and denoted by $\delta_s = \sqrt{\sum_{n=1}^{N_{in}} (\delta_n^s - e_s)^2 / N_{in}}$ and $\delta_{pX} = \sqrt{\sum_{n=1}^{N_{in}} (\delta_n^{pX} - e_{pX})^2 / N_{in}}$ respectively. In the experiment, we have $N_{in} = 36$ for which we vary $\varphi_n$ from $0^\circ$ to $350^\circ$ by every $10^\circ$ and the probed time window of 7 picoseconds is divided into $N_T = 106$ points for data acquisition. From the experimental data and the above procedure, we get $e_s = 4.2\%$, $e_{pS} = 7.8\%$, $e_{pA1} = 5.9\%$, and $e_{pA2} = 3.2\%$ and $\delta_s = 2.3\%$, $\delta_{pS} = 3.3\%$, $\delta_{pA1} = 2.6\%$ and $\delta_{pA2} = 1.6\%$. The fitting to the S-wave data by mirror-symmetric function $E_{em}^s(t, \varphi) = f_S^a(t, \varphi)$ is thus plausible while the best fitting here to the P-wave data is the
mirror-asymmetric function given by $f_{\perp}^{\parallel}(t, \phi) = f_{\perp}^{\parallel}(t, \phi)$. The electric field component $E_{em}^{\parallel}(t, \phi)$ is then extracted accordingly by $E_{em}^{\parallel}(t, \phi) = f_{\perp}^{\parallel}(t, \phi) - f_{\perp}^{\parallel}(t, \phi)$. Hereby we have supplemented details of the fitting functions $E_{em}^{\parallel}(t, \phi)$ and $E_{em}^{\parallel}(t, \phi)$ mentioned in Sec. 2 above.

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By assuming the smallness of $\delta t$, the value of $P(t,t')f(t')$ becomes non-negligible only for $t'$ near $t$ such that $\tau_{k_{n'}}$ is approximately by $\tau_{k_{n}}$.

Conventionally, the second-order current is found by solving the Boltzmann equation for one band $n$, namely, 
\[
\frac{\partial f_n(k_{n},t)}{\partial t} + \langle \mathbf{E}(t) \cdot \mathbf{v}(k_{n},t) \rangle / \tau_{k_{n}} = -\langle f_n(k_{n},t) - f_n^{eq}(k_{n}) \rangle / \tau_{k_{n}} \quad \text{with} \quad f_n(k_{n},t) \quad \text{considered up to the second order in the electric field and one computes the current by} \quad j(t) = -e \int d^2 k f_n(k, t) v_n(k) \quad \text{where} \quad v_n(k) \quad \text{is given by the right-hand side of Eq. (A18). Following this approach with the partial application of the short-relaxation time approximation Eq. (A8), that leaves the form of} \quad \mathbf{E}(t') \quad \text{intact in the relevant time integral, one then obtains the result of Eq. (A19).}
\]

These results are obtained by solving $(\hbar/2) d(\langle \mathbf{\sigma}_{n,-}(k_{n},t) \rangle / dt = \Lambda_{so}(k_{n}) \times \langle \mathbf{\sigma}_{n,-}(k_{n},t) \rangle$, as a result of $i\hbar \left\{ \phi(k_{n},t), t_0 \right\} = \mathcal{H}(k_{n}) \left\{ \phi(k_{n},t), t_0 \right\}$, from the initial texture $\langle \mathbf{\sigma}_{n,-}(k_{n},t) \rangle \mid _{t_0} = \langle \mathbf{\sigma}_{n,-}(k_{n}) \rangle$, to a later time $t = t_0 + \delta t$ using the fourth order Runge-Kutta method with sufficiently small $\delta t$. Here in Eq. (C1), we have $\delta \sigma_{ij} = -(2/3) \left[ \alpha^2 R_{K_{t1/2}} \left( K_{t1} + K_{t1/2} \right) k \right] (\delta t / \hbar)^2$ and $\delta \phi_{ij} = -(2/3) \left[ \alpha^2 R_{K_{t1/2}} \left( K_{t1} + K_{t1/2} \right) k \right] (\delta t / \hbar)^3$. They are dimensionless amplitudes independent of $\theta$ and $\varphi_{\phi}$ in which $K_{t} = |K_{t}|$ and $t_{1/2} = t_0 + \delta t / 2$.

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This result is seen from Eq. (C1) where one finds $\int_0^{\pi} d\theta \langle \sigma_{||/\perp}(k_{1},t_1) \rangle \propto \int_0^{\pi} d\theta \Gamma_{1}(x_{||/\perp}, \theta, \varphi_{\phi}) = \int_0^{\pi/4} d\theta \left( x_{||/\perp} + x_{||/\perp} \right) \cos(2\theta) + \left( x_{||/\perp} - x_{||/\perp} \right) \sin(2\theta) / \sqrt{r_A^2 \cos^2 \theta + r_A^2 \sin^2 \theta}$ where $x_{||/\perp} = \sqrt{v_A^2}$ and $x_{||/\perp} = r_A$ by using that the function $\sin(2\theta) / \sqrt{r_A^2 \cos^2 \theta + r_A^2 \sin^2 \theta}$ is odd in the range of integration.

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