Unblocking time-reversal forbidden photocurrents in non-magnetic materials

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Magnetic materials enable to access a wide set of responses not available when time-reversal symmetry is preserved. A prime example are time-reversal forbidden photocurrent responses (e.g., linear injection and circular shift photocurrents) that are thought to vanish in non-magnetic materials; such photocurrent responses can, instead, manifest in parity violating magnets exhibiting a current that is controlled by magnetic order. Here we argue that non-vertical interband transitions enable to unblock such time-reversal forbidden photocurrents even in non-magnetic and inversion symmetric materials, and can display a rich phenomenology distinct from their vertical counterparts. Strikingly, these non-vertical photocurrents exhibit a resonant peak-like structure when photoexcited carriers are close to the Fermi surface. Arising from a cooperative effect between finite \( q \) irradiation and the Fermi surface position, these non-vertical transitions enable to selectively address parts of the Fermi surface to yield giant enhancements of photocurrent. Realization of such non-vertical time-reversal forbidden photocurrents can enable a powerful photocurrent tool to interrogate the quantum geometry of metals even when time-reversal and inversion symmetries remain intact.

Time-reversal (\( T \)) symmetry plays a central role in determining the responses of quantum materials. A prominent example is how \( T \) symmetry forbids elastic backscattering in topological helical edge states allowing to realize a quantized edge conductance \([1,3]\). Breaking \( T \) symmetry is essential in realizing Hall currents and can enable access to a host of quantum geometric quantities such as Berry curvature \([4]\) and the quantum metric \([5,7]\). No less striking is the impact of \( T \) symmetry on nonlinear optical responses. Of particular interest are the generation of nonlinear bulk photocurrents (e.g., shift and injection photocurrents) that can arise in non-centrosymmetric materials even in the absence of a \( p-n \) junction \([8,15]\), see Fig. 1a, b. For instance, linear shift and circular injection photocurrents are allowed in \( T \) invariant materials \([11,13]\). In contrast, due to their odd-parity under \( T \), circular shift (CS) and linear injection (LI) photocurrents are forbidden in \( T \) invariant non-magnetic materials \([13,14]\). As a result, CS/LI photocurrents (time-reversal forbidden photocurrents) are thought to only manifest in parity violating magnetic materials \([13,15]\) such as antiferromagnets.

Here we argue that non-vertical interband transitions enable to unblock time-reversal forbidden photocurrents even in non-magnetic and inversion (\( P \)) symmetric materials, Fig. 1c. In particular, we find that electromagnetic (EM) irradiation with a finite wavevector \( q \) \([10]\) induce non-vertical bulk CS and LI photocurrents (time-reversal forbidden photocurrents) that have a rich phenomenology distinct from that of conventional bulk photovoltaic currents at \( q = 0 \) (see e.g., Table 1). Strikingly, we find that non-vertical CS/LI photocurrents exhibit an unusual Fermi surface resonance (FSR) like structure yielding large photocurrent peaks when EM irradiation photoexcites carriers close to the Fermi surface (see e.g., Fig. 2 for the non-vertical CS photocurrent); the photocurrent becomes suppressed when tuned away from the FSR. This contrasts starkly with that typically expected of conventional vertical bulk photovoltaic currents (e.g., circular injection and linear shift photocurrent) that can display large responses far away from the Fermi surface \([10,12]\). As we will discuss below, the FSR effect enables large non-vertical CS/LI photocurrents to be produced even for modest values of momentum transfer.
We anticipate that non-vertical time-reversal forbidden (CS/LI) photocurrents and their FSR behavior can be readily realized in a wide variety of $T$ and $P$ symmetric materials. Even as we expect non-vertical bulk photovoltaic currents can be realized in bulk three-dimensional (3D) materials with both $T$ and $P$ symmetries (e.g., 3D Dirac semimetals [17]), of special interest, however, are two-dimensional (2D) materials. In these, exposed 3D Dirac semimetals [17]), of special interest, however, can be realized in bulk three-dimensional materials. Even as we expect non-vertical bulk photocurrents and their FSR behavior can be readily realized in a wide variety of $T$ and $P$ symmetric materials. Even as we expect non-vertical bulk photovoltaic currents can be realized in bulk three-dimensional (3D) materials with both $T$ and $P$ symmetries (e.g., 3D Dirac semimetals [17]), of special interest, however, are two-dimensional (2D) materials. In these, exposed 

Non-vertical injection and shift photocurrents – We begin with a Hamiltonian describing the interaction between an electronic material with incident finite-q electromagnetic (EM) irradiating fields [16]:

$$\mathcal{H} = \mathcal{H}^{(0)} + \mathcal{V}, \quad \mathcal{V} = -\frac{e}{2\epsilon} [\mathbf{A}(r,t) \cdot \mathbf{v}^{(0)} + \mathbf{v}^{(0)} \cdot \mathbf{A}(r,t)]$$ (1)

where $\mathcal{H}^{(0)}(r)$ is the bare Hamiltonian of the material, and $\mathcal{V}(r,t)$ describes the finite wavevector light-matter interaction [16]. Here $\mathbf{A}(r,t)$ is a spatially varying vector potential that captures the finite wavevector EM fields of the incident radiation, $e$ is the carrier charge, $\mathbf{v}^{(0)}$ is the velocity operator, and $c$ is the speed of light. In what follows, we will consider incident finite-q EM fields with the real-valued electric field profile $\mathbf{E}(r,t) = (1/2) \sum_{\pm} E_{\pm} e^{i \mathbf{q} \cdot \mathbf{r} \mp i \omega t}$ with $E_{\pm}$ the complex electric field amplitude where $E_{\pm} = (E_{\mp})^*$. The oscillating EM fields induce real electronic transitions between states $\{n,p\}$ and $\{m,p'\}$ with rate captured by Fermi’s golden rule:

$$W_{n,p \rightarrow m,p'}^{\pm} = \frac{2\pi}{\hbar} |M_{mn}^{\pm}|^2 \delta^{\pm} f(e_{n,p}) \left[1 - f(e_{m,p'})\right]$$ (2)

where $\delta^{\pm} = \delta(e_{n,p'} - e_{n,p} \mp \hbar \omega)$ satisfies energy conservation, and the matrix element can be obtained from Eq. (1) as $|M_{mn}^{\pm}|^2 = (e^2/4\omega^2) \cdot |\langle m,p | E_{\pm} \cdot \mathbf{v}^{(0)} | n,p \rangle|^2$. The latter imposes momentum conservation and we have used $\partial_t \mathcal{A}(r,t) = -e \mathbf{E}(r,t)$. Here $f(e)$ are Fermi functions and $\mathbf{v}(k) = \partial \mathcal{H}(k)/\hbar \partial \mathbf{k}$ where $\mathcal{H}(k)$ is the Bloch Hamiltonian. When $\mathbf{q} = 0$ (e.g., for normal incident far-field light), vertical interband transitions take place. However, when $\mathbf{q} \neq 0$ (e.g., when coupled to plasmonic modes), non-vertical transitions ensue [16]. As we will see below, such non-vertical transitions unblock time-reversal forbidden nonlinear photocurrents.

To see this, we first examine the injection current that arises from a change of velocity when a carrier transits from an initial state $i$ to a final state $f$ [8, 13]:

$$\partial_t j^{\text{inj}}(q) = e \sum_{i \rightarrow f} W_{i \rightarrow f} \Delta_{i \rightarrow f}, \quad \Delta_{i \rightarrow f} = v_f - v_i$$ (3)

where $\mathbf{v}_{i,f}$ are the velocities of the initial and final state respectively. As a concrete demonstration and for clarity and brevity of presentation, we focus on a two-band system where EM radiation induces transitions between the conduction $c$ and the valence $v$ bands. The same analysis, discussed below, can be extended for general multi-band systems by summing across pairs of transitions. Considering momentum and energy conservation, EM radiation induces transitions between pairs of Bloch states $|u_c(k - q/2)\rangle$ and $|u_v(k + q/2)\rangle$, where $W^\pm$ are associated with $v \rightarrow c$, and $W^-\mp$ with $c \rightarrow v$ [30]; here $k$ is the Bloch wavevector. Summing over all transitions in Eq. (3) and using the rates in Eq. (2), we obtain the finite-q injection current:

$$\partial_t j^{\text{inj}}(q) = C \sum_k \rho(k,q) |\mathbf{E} \cdot \mathbf{v}_{cv}(k,q)|^2 \Delta(k,q),$$ (4)

where $C = -e^3 \pi/(2\hbar^3)$, the occupation factor $\rho(k,q) = f_{cv}(k,q) \delta(\epsilon_{cv}(k,q) - \hbar \omega)$, the Fermi function difference is $f_{cv}(k,q) = f_{cv}(k,q + \mathbf{q}/2) - f_{cv}(k,q - \mathbf{q}/2)$ with the interband transition energy $\epsilon_{cv}(k,q) = \epsilon_c(k + q/2) - \epsilon_v(k - q/2)$, the velocity matrix element is $\mathbf{v}_{cv}(k,q) = |\langle u_v(k + q/2) | \mathbf{v}(k) | u_c(k - q/2) \rangle|$, electric field amplitude $\mathbf{E} = E^\| \mathbf{k}$, and $\Delta(k,q) = v_c(k + q/2) - v_v(k - q/2)$ is the change in carrier velocity. When $\mathbf{q} = 0$, Eq. (4) reduces to the well-known expression for (vertical) injection current [13].

In the same fashion, the shift current [8, 9, 13] can be expressed by instead accounting for the real-space displacements of carriers as they transition from $i \rightarrow f$ [13]. Using Eq. (2), the finite-q shift current is [20]

$$j^{\text{shift}}(q) = C \sum_k \rho(k,q) |\mathbf{E} \cdot \mathbf{v}_{cv}(k,q)|^2 r(k,q),$$ (5)

where $r(k,q)$ is the real-space displacement [10, 13, 21] when a valence electron transits to the conduction band $r(k,q) = A_v(k + q/2) - A_v(k - q/2) - \nabla k \arg \left| \mathbf{E} \cdot \mathbf{v}_{cv}(k,q) \right|$, with $A_v(k) = (u_v(k) | i \nabla_k | u_v(k))$ the Berry connection.

Unblocking time-reversal forbidden photocurrents – As we now argue, both $j^{\text{inj}}(q)$ and $j^{\text{shift}}(q)$ in Eq. (4) and Eq. (5) depend on four quantities: $\rho(k,q)$ and $\Delta(k,q)$ as well as $\mathbf{E} \cdot \mathbf{v}_{cv}(k,q)$ and $r(k,q)$. While the first two are intrinsic bandstructure quantities that are $\mathbf{E}$ polarization independent, the latter two depend on $\mathbf{E}$ polarization. We will analyze each in turn.

We proceed by noting that the band energies are even under either $T$ or $P$-symmetry. As a result, the non-vertical occupation factor $\rho(k,q)$ as well as the velocity
TABLE I: Symmetry relations for non-vertical charge shift and injection photocurrents. Photocurrents for linear polarized irradiation are denoted \( \theta \) whereas helicity dependent photocurrents for circularly polarized irradiation are denoted “cir”, see text. We find that non-vertical \( \mathbf{q} \neq 0 \) LI and CS photocurrents are allowed in both \( T \) and \( P \)-preserving materials (indicated by ticks, third row). In contrast, when \( \mathbf{q} = 0 \) LI and CS photocurrents are time-reversal forbidden in \( T \)-preserving but \( P \)-breaking materials. The situation is flipped for circular injection and linear shift photocurrents which vanish even for non-vertical \( \mathbf{q} \neq 0 \) (third row) in \( T \) and \( P \)-preserving materials, but are allowed when \( \mathbf{q} \neq 0 \) for \( T \)-preserving but \( P \)-breaking materials. Naturally, when \( P \) symmetry remain unbroken, all \( \mathbf{q} = 0 \) nonlinear photocurrents vanish identically; similarly, when either \( P \) or \( T \) symmetry, all \( \mathbf{q} \neq 0 \) non-linear photocurrents are allowed.

| charge photocurrent | linear injection | circular injection | linear shift | circular shift |
|---------------------|------------------|-------------------|-------------|---------------|
| \( P \)-symmetry    | \( \partial_t \mathbf{j}_\text{L}(\mathbf{q}) = -\partial_t \mathbf{j}_\text{L}(\mathbf{q}) \) | \( \partial_t \mathbf{j}_\text{L}(\mathbf{q}) = -\partial_t \mathbf{j}_\text{L}(\mathbf{q}) \) | \( j_\text{L}(\mathbf{q}) = -j_\text{L}(\mathbf{q}) \) | \( j_\text{L}(\mathbf{q}) = -j_\text{L}(\mathbf{q}) \) |
| \( T \)-symmetry    | \( \partial_t \mathbf{j}_\text{L}(\mathbf{q}) = -\partial_t \mathbf{j}_\text{L}(\mathbf{q}) \) | \( \partial_t \mathbf{j}_\text{L}(\mathbf{q}) = \partial_t \mathbf{j}_\text{L}(\mathbf{q}) \) | \( j_\text{L}(\mathbf{q}) = j_\text{L}(\mathbf{q}) \) | \( j_\text{L}(\mathbf{q}) = -j_\text{L}(\mathbf{q}) \) |

\( \mathbf{q} \neq 0 \ (T \& \ P \text{ symmetry}) \) \( \checkmark \) \( \times \) \( \times \) \( \checkmark \)

\( \mathbf{q} = 0 \ (T \text{ symmetry only}) \) \( \times \) \( \checkmark \) \( \checkmark \) \( \times \)

\( \rho(\mathbf{k}, \mathbf{q}) = \rho(-\mathbf{k}, -\mathbf{q}), \ \Delta(\mathbf{k}, \mathbf{q}) = -\Delta(-\mathbf{k}, -\mathbf{q}), \ \ (7) \)

under either \( T \) or \( P \)-symmetry. Here have noted that \( \Delta(\mathbf{k}, \mathbf{q}) = \nabla_k \epsilon_{\text{ev}}(\mathbf{k}, \mathbf{q}).\)

We now turn to analyzing the polarization dependent quantities. To do so, it is useful to denote linearly polarized \( \mathbf{E}^l = \mathbf{E}_0(\cos \theta \mathbf{x} + \sin \theta \mathbf{y}) \) and circularly polarised \( \mathbf{E}^c = \mathbf{E}_0(\mathbf{x} + i\mathbf{y}) \), where \( \eta = \pm 1 \) electric fields by the index \( \theta(\eta) \). By accounting for how \( T \) or \( P \)-symmetries transform the interband velocity matrix element \( \mathbf{SI} \), we find the square of the interband transition matrix element \( \eta^{(\eta)}(\mathbf{k}, \mathbf{q}) = |\mathbf{E}^\theta(\eta) \cdot \mathbf{\nu}_{\text{ev}}(\mathbf{k}, \mathbf{q})|^2 \) obeys:

\( T \)-symmetry : \( \eta^{\theta(\eta)}(\mathbf{k}, \mathbf{q}) = \eta^{\theta(-\eta)}(-\mathbf{k}, -\mathbf{q}), \)

\( P \)-symmetry : \( \eta^{\theta(\eta)}(\mathbf{k}, \mathbf{q}) = \eta^{\theta(-\eta)}(-\mathbf{k}, -\mathbf{q}), \)

where we have recalled that for a given \( \eta \) polarization, the anti-unitary nature of \( T \) acting on \( \mathbf{\nu}_{\text{ev}}(\mathbf{k}, \mathbf{q}) \) effectively flips the helicity of the light irradiation in the interband transition rate \( \eta^\theta \), see \( \mathbf{SI} \).

Similarly, the \( \eta \) dependent generalized shift vector in \( \mathbf{E} \) also depends on \( \mathbf{E} \) polarization; in the same fashion as above we will index the generalized shift vector for linear (circular) polarized \( \mathbf{E} \) irradiation by \( \eta(\theta) \): \( \eta^{(\theta)}(\mathbf{k}, \mathbf{q}) \).

Similar to \( T \) and \( P \) symmetries, the shift vectors obey:

\( T \)-symmetry : \( \eta^{\theta(\eta)}(\mathbf{k}, \mathbf{q}) = \eta^{\theta(-\eta)}(-\mathbf{k}, -\mathbf{q}), \)

\( P \)-symmetry : \( \eta^{\theta(\eta)}(\mathbf{k}, \mathbf{q}) = -\eta^{\theta(\eta)}(-\mathbf{k}, -\mathbf{q}), \)

where we have accounted for how the geometric phase accru during the interband transition transforms \( \mathbf{SI} \).

Applying the symmetry relations in Eq. \((6) \) to the non-vertical linear (circular) injection and shift photocurrents in Eq. \((1) \) and Eq. \((5) \), and summing across all \( \mathbf{k} \) yields the non-vertical photocurrent symmetry properties shown in Table 1 (see e.g., first two rows). In populating the table, we have denoted photocurrents arising from linear polarized \( \mathbf{E} = \mathbf{E}^l \) with the subscript index \( \theta \). Similarly, to isolate the photocurrent that depends on the helicity of light for circularly polarized irradiation \( \mathbf{E} = \mathbf{E}^c \), we have focussed on non-vertical circular injection currents \( [\partial_t \mathbf{j}_\text{cir}(\mathbf{q}) = \sum_\eta \eta \partial_t \mathbf{j}_\text{cir}^\eta(\mathbf{q})] \) and non-vertical circular shift photocurrents \( [\mathbf{j}^\text{shift(\eta)} = \sum_\eta \eta \mathbf{j}^\text{shift(\eta)}] \), denoted with the subscript “cir”.

Of particular note are the LI and CS photocurrents (first and fourth columns). Even as both of these photocurrents are (time-reversal) forbidden when \( \mathbf{q} = 0 \) in \( T \) invariant non-magnetic materials (fourth row), non-vertical transitions when \( \mathbf{q} \neq 0 \) enable to generate finite non-vertical LI and CS photocurrents even in materials with both \( T \) and \( P \) symmetries (third row). This is because LI/CS photocurrents display an odd parity as \( \mathbf{q} \rightarrow -\mathbf{q} \) for either \( T \) and \( P \) symmetries: \( \mathbf{q} \) controls the direction of the non-vertical LI/CS photocurrent generated in such \( T \) and \( P \) symmetric materials.

This behavior contrasts starkly with that of the circular injection and linear shift photocurrents (second and third columns). While both these photocurrents are allowed when \( \mathbf{q} = 0 \) in \( T \) invariant non-magnetic materials (fourth row), non-vertical circular injection and linear shift photocurrents possess an even parity when \( \mathbf{q} \neq -\mathbf{q} \) in materials with \( T \) symmetry. As a result, non-vertical circular injection and linear shift photocurrents vanish in materials possessing both \( T \) and \( P \) symmetries – non-magnetic centrosymmetric materials. This dichotomy between LI/CS photocurrents and the circular injection and linear shift photocurrents found for \( \mathbf{q} \neq 0 \) (row 3, non-vertical) and \( \mathbf{q} = 0 \) (row 4, vertical) closely mirrors that of a similar delineation found for vertical photocurrents between \( \mathcal{PT} \) symmetric parity-violating magnets and \( T \)-invariant non-centrosymmetric materials recently discussed in Ref. \([13, 14]\).

When either \( T \) or \( P \) symmetry are broken, all non-vertical injection and shift photocurrents are allowed. In-
terestingly, we note that the (contrasting) odd-nature of LI/CS photocurrents when \( \mathbf{q} \rightarrow -\mathbf{q} \) under time-reversal symmetry, enables to identify them even in \( \mathcal{P} \) breaking (but \( \mathcal{T} \)-preserving) systems. Lastly, we note that while we have concentrated on charge photocurrent response, non-vertical spin photocurrents are expected to have different transformation properties from that of Table 1. In particular, we note that non-vertical interband transition processes can also give rise non-vertical spin photocurrents even in \( \mathcal{P} \) and \( \mathcal{T} \) preserving materials [20].

**Non-vertical CS and LI photocurrents in bilayer graphene and Fermi surface resonance** — As a concrete demonstration of how non-vertical transitions unblock time-reversal forbidden photocurrents, we examine CS and LI photocurrents in gapless bilayer graphene. Notably, bilayer graphene is a centrosymmetric semimetal that preserves \( \mathcal{T} \)-symmetry; all \( \mathbf{q} = 0 \) bulk photovoltaic effects vanish. Its low energy Hamiltonian can be written as

\[
H(\mathbf{p}) = H_0(\mathbf{p}) + H_w(\mathbf{p})
\]

where

\[
H_0(\mathbf{p}) = -\frac{\hbar^2}{2m} \left[ \left( p_x^2 - p_y^2 \right) \sigma_x + 2\zeta p_x p_y \sigma_y \right],
\]

\[
H_w(\mathbf{p}) = \hbar v_f (\zeta p_x \sigma_x - p_y \sigma_y).
\]

Here \( \mathbf{p} = \mathbf{k} - \mathbf{K}_\zeta \) is the Bloch wavevector measured from \( K_\zeta \) points, \( \zeta = \pm \) is the valley index, and \( m \) is the effective mass. \( H_w(\mathbf{p}) \) describes trigonal warping.

We first examine the non-vertical CS photocurrent. We numerically evaluate Eq. (5) for a circularly polarized beam (\( \hbar \omega = 200 \text{ meV} \)) with in-plane photon wavevector \( q || \hat{y} \) along a mirror axis in bilayer graphene. This yields a sizeable non-vertical \( J_{\text{shift}} \) in Fig. 2 in \( \mathcal{P} \) and \( \mathcal{T} \) preserving bilayer graphene as expected from Table 1. We note that \( J_{\text{shift}} \) shown is purely transverse to \( q \) flowing along \( \hat{x} \); this behavior arises due to the mirror axis along \( \hat{y} \) (SI).

Strikingly, non-vertical circular shift current display large peaks centered at \( \mu = \pm \hbar \omega/2 \) (shaded regions in Fig. 2b). These Fermi surface resonant (FSR) peaks arise that reign bilayer graphene as expected from Table 1. We note that \( J_{\text{shift}} \) shown is purely transverse to \( q \) flowing along \( \hat{x} \); this behavior arises due to the mirror axis along \( \hat{y} \) (SI).

To understand this physically, we plot the distribution of the weighted shift vector \( \mathbf{R}^{\zeta,\eta}(\mathbf{p}, \mathbf{q}) = v^{\zeta,\eta}(\mathbf{p}, \mathbf{q}) \delta(\mathbf{p}, \mathbf{q}) \) [this determines the direction of the non-vertical CS photocurrent, see Eq. (5)] in Fig. 2b and c. Here the interband transition contours (black) indicate \( \mathbf{p} \) values that satisfy \( \delta(\epsilon_{cv}(\mathbf{p}, \mathbf{q}) - \hbar \omega) \). When \( \mu = -\hbar \omega/2 \) (in the valence band), the Fermi surface intersects with the interband transition contour so that only the bottom half of the transition contour contributes to the non-vertical interband transitions (solid curve in Fig. 2b). These \( \mathbf{p} \) values correspond to occupied carriers in the valence band so that \( f_{cv}(\mathbf{p}, \mathbf{q}) \neq 0 \). In contrast, the other half (dashed curve) do not contribute to the non-vertical interband transitions \( (f_{cv}(\mathbf{p}, \mathbf{q}) = 0) \). This asymmetric sampling (enforced by the occupation factors) of the interband transition contour enables large non-vertical CS photocurrents to accrue. Similarly, when \( \mu = \hbar \omega/2 \) is in the conduction band (Fig. 2b), only the top half of the transition contour is available for interband transitions.

![FIG. 2: Non-vertical charge CS photocurrent in T- and P-symmetric bilayer graphene. (a) Non-vertical CS photocurrent as a function of Fermi energy \( \mu \). Blue, orange, yellow and purple curves are obtained at temperatures 10 K, 20 K, 50 K and 100 K. (b,c) Fermi surface resonance arises from the imbalanced sampling of shift vector when carriers close to the Fermi surface are excited. Weighted non-vertical circular shift vector for \( \eta = +1 \) with contour line plots (black) indicating the regions that satisfy energy and momentum conservation. Solid line indicate shift vector regions that are sampled, dashed indicate regions that are not sampled when chemical potential is fixed at \( \mu = -\hbar \omega/2 \) (b) and \( \mu = \hbar \omega/2 \) (c) in the \( K_+ \) (top panel) and \( K_- \) (bottom panel) valleys. Parameters used: \( \hbar \omega = 200 \text{ meV}, |q| = 0.001 \text{ nm}^{-1} \) (corresponding to the free-space wavelength), and electric field amplitude 0.05 V/\( \mu \text{m} \). Here the wavevector \( \mathbf{q} \) is directed along \( \hat{y} \) yielding a transverse current; when \( \mathbf{q} \) direction is reversed, the sign of \( J_{\text{shift}} \) flips, see Table 1.](image-url)
Temperature dependence of the non-vertical LI photocurrent need to be more tilted to intersect with the Fermi surface. (b) 200 meV and the electric field amplitude is $E$ an FSR jing the directions of the weighted shift vector $R^{c,v}(p, q)$ along the interband transition contour. Interestingly, the Hamiltonian in Eq. (10) possesses an emergent $c, v$ band symmetry that enforces an even $c^{cv}_p(p, q) = c^{cv}_p(-p, q)$ but an odd $R^{c,v}(p, q) = -R^{c,v}(-p, q)$ (see vector plots in Fig. 2b,c), see also SI. As a result, in this regime when summing across the entire transition contour, non-vertical $j^\text{shift}$ vanishes. We note $c, v$ symmetry can lead to a non-vanishing $j^\text{shift}$ away from FSR. For bilayer graphene, such $c, v$ asymmetry, however, is small and results in non-vertical $j^\text{shift}$ that are significantly suppressed away from FSR.

As we will now see, FSR is a ubiquitous feature of non-vertical photocurrents. Indeed, we find FSR peaks also appear for the non-vertical LI photocurrent. In Fig. 3 we find large $\partial_{\nu}j^{\text{inj}}$ (where we used $E = E_0 \hat{y}$ and $q \parallel \hat{E}$) in $\mathcal{T}$ and $P$ preserving bilayer graphene as expected from Table 1. Due to mirror symmetry, when $q$ is aligned with the mirror axis, the non-vertical charge LI photocurrent is entirely longitudinal. Similar to $j^\text{shift}$ discussed above, $\partial_{\nu}j^{\text{inj}}$ peaks in the FSR regime and is suppressed when $\mu$ moves away from the FSR regime. Such a resonant-peak-like structure can also exist in monolayer graphene as well where a similar photon-drag photocurrent for oblique incident far-field linearly polarized light has been predicted at low-temperature [23].

Coupling to polaritons in the near-field (e.g., via plasmon or other polaritons), however, allows to use a far larger range of $q$. In 2D plasmonic heterostructures [18, 19], wavelength compression of up to several hundred times can be achieved [18, 19] As we now show, this enables to realize non-vertical LI photocurrents at high temperatures as well as access a rich phenomenology of non-vertical transitions. Indeed, we find that when $\mu = -\hbar\omega/2$, non-vertical $\partial_{\nu}j^{\text{inj}}$ rapidly rises with $q$ (blue curve in Fig. 3b) before plateauing at larger $q$ values. Strikingly, small $q$ valued non-vertical $\partial_{\nu}j^{\text{inj}}$ are highly temperature sensitive and fall quickly as temperature increases (see blue solid curve, Fig. 3b for $q = 0.01 \text{ nm}^{-1}$); in contrast, when $q$ is large (see dashed blue Fig. 3b) for $q = 0.1 \text{ nm}^{-1}$, $\partial_{\nu}j^{\text{inj}}$ becomes less temperature sensitive, allowing significant photocurrents to persist to high temperatures.

When $\mu$ is slightly tuned away from $\pm\hbar\omega/2$ (orange and purple curves Fig. 3b), the FSR effect described above does not kick in immediately. Indeed at small $|q|$, non-vertical linear injection photocurrents vanishes for $\mu$ tuned far away from $-\hbar\omega/2$ due to the presence of $c, v$ band symmetry in Eq. (10), in a similar fashion to that discussed above for the non-vertical CS photocurrents, see also SI. We note, parenthetically, that when $c, v$ bands are asymmetric, small but $(c, v$ band asymme-
try dependent) non-vanishing photocurrents for linearly polarized incident irradiation can be induced even when \( \mu \) is tuned away from \( -\hbar \omega/2 \), see Ref. [24] for an example in graphene. When \( |q| \) exceeds a threshold value, however, the transition contour is significantly tilted and starts to intersect with the Fermi surface (see rapid turn-on region for orange and purple curves) activating FSR non-vertical \( \partial j_0^{\text{inj}} \).

Interestingly, for these \( \mu \) values tuned away from FSR, non-vertical \( \partial j_0^{\text{inj}} \) display a \( q \) dependent temperature dependence. For instance, for small \( q \) (e.g., black star), the non-vertical \( \partial j_0^{\text{inj}} \) for \( \mu \) away from \( -\hbar \omega/2 \) increase as temperature increases (orange and purple curves Fig. [3a,b]). This temperature dependence can be understood by examining the parts of the interband transition contour that contributes to photocurrent: for e.g., when \( \mu \) is away from \( -\hbar \omega/2 \) (Fig. [3]), as temperature increases, the part of the transition contour with lower energy starts to get more occupied due to thermal excitation thereby increasing non-vertical \( \partial j_0^{\text{inj}} \) (Fig. [3]). The situation is distinctly different when \( q \) is large (e.g., blue star). Here large swaths of the interband transition contour are already occupied and display near optimal non-vertical \( \partial j_0^{\text{inj}} \) at low temperature (Fig. [3]); increasing temperature in this regime further expands the region of the interband transition contour that contributes to \( \partial j_0^{\text{inj}} \) (Fig. [3], see faint yellow \( f_{cv} \)): partial cancellation of \( \Delta^+(p,q) \) in \( k \)-space leads to a decreasing current (dashed orange curve in Fig. [3]).

Non-vertical processes can enable to realize time-reversal forbidden photocurrents in non-magnetic (and inversion symmetric) systems, enabling to probe time-reversal forbidden (LI and CS) photocurrents in a wide range of materials where either \( P \) or \( T \) or both symmetries are intact. The large resonant-like peaks when carriers close to the Fermi surface are excited provide giant enhancements to non-vertical photocurrents. Strikingly, FSR also provides means to selectively address parts of the Fermi-surface, with an angular resolution that can be controlled by the \( q \) wavevector of the incident EM radiation that is reminiscent of angular-resolved photoemission [25]. We note that nonlinear photocurrents depend intimately on the quantum geometry of materials; e.g., the LI photocurrents of Eq. [1] depend on the interband quantum metric associated with the transition [13]. Since the FSR addresses selective parts of the Fermi surface (see e.g., Fig. [3]), such non-vertical photocurrents may enable new means to study momentum-resolved quantum geometry on the Fermi surface [14] [26] [27]; it may also allow to excite collimated beams of primary photoexcited carriers that can be used to probe their relaxation kinematics [28] [29].

From a technological perspective, we anticipate that the FSR regime allows to realize large nonlinear susceptibilities. Taking the non-vertical injection current [where \( \partial j_0(q) = \eta_{abc}(q)\epsilon_0(q)E^2(q) \)] as an example, we estimate susceptibilities as high as \( \eta_{abc}(q) \approx 10^{10} - 10^{11} \text{A mm}^{-2} \text{V}^{-2} \text{s}^{-1} \) can be achieved in bilayer graphene (a \( P \) and \( T \) preserving material); these values are comparable to those recently discussed for \( \eta_{xyz}(q = 0) \) (for vertical circular injection photocurrent) found in 2D ferroelectric materials [12].

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Supplementary Information for “Unblocking time-reversal forbidden photocurrents in non-magnetic materials”

I. Symmetry Analysis for Non-vertical Shift and Injection Currents

In this section, we discuss the symmetry properties of photon drag shift and injection currents induced by linearly or circularly polarised light. We will demonstrate that properties of the non-vertical injection and shift photocurrents are sensitive to the symmetry of the material irradiated as well as the light polarisation. These properties can be obtained by examining how the Bloch wavefunction and velocity matrix elements transform under various symmetry operators.

We begin with the Bloch Hamiltonian \( H(k) = e^{-i\mathbf{k} \cdot \mathbf{r}} H(r)e^{i\mathbf{k} \cdot \mathbf{r}} \). The Bloch wavefunction \( |u_n(k)\rangle \) in band \( n \) satisfies \( H(k)|u_n(k)\rangle = \epsilon_{n,k}|u_n(k)\rangle \). We proceed by considering how the Bloch hamiltonian and its associated Bloch wavefunctions transform when the material possesses (i) spatial inversion (\( P \)) operators.

obtained by examining how the Bloch wavefunction and velocity matrix elements transform under various symmetry

are sensitive to the symmetry of the material irradiated as well as the light polarisation. These properties can be

yielding the following constraints on the energy dispersion and the Bloch wavefunctions:

\[
\epsilon_{n,k} = \epsilon_{n,-k}, \quad \mathcal{P}|u_n(k)\rangle = C_{n,k}|u_n(-k)\rangle,
\]

where \( C_{n,k} \) is a complex phase factor associated with the \( \mathcal{P} \) transformation satisfying \( |C_{n,k}| = 1 \). Since \( \mathcal{P} \) is unitary and preserves inner product, we have

\[
\langle u_n(-k_1)|u_m(-k_2)\rangle = (C_{n,k_1}^* \mathcal{P} u_n(k_1)|C_{m,k_2}^* \mathcal{P} u_m(k_2)) = C_{n,k_1} C_{m,k_2}^* \langle u_n(k_1)|u_m(k_2)\rangle.
\] (S3)

Furthermore, under spatial inversion symmetry, the velocity operator transforms as \( \mathcal{P} \hat{\nu} \mathcal{P}^{-1} = -\hat{\nu} \). Thus the velocity matrix element satisfies

\[
\langle u_n(k_1)|\hat{\nu}|u_m(k_2)\rangle = \langle u_n(k_1)|\mathcal{P}^{-1} \hat{\nu} \mathcal{P}^{-1}|u_m(k_2)\rangle = -C_{n,k_1}^* C_{m,k_2}^* \langle u_n(-k_1)|\hat{\nu}|u_m(-k_2)\rangle.
\] (S4)

In the same fashion as above, when the material possesses time-reversal symmetry, the energy dispersion and Bloch wavefunctions transform as

\[
\epsilon_{n,k} = \epsilon_{n,-k}, \quad \mathcal{T}|u_n(k)\rangle = C_{n,k}'|u_n(-k)\rangle,
\]

where \( C_{n,k}' \) is a complex phase factor associated to \( T \) operation with \( |C_{n,k}'| = 1 \). In addition, since \( \mathcal{T} \) is anti-unitary, the inner product of the wavefunctions satisfies

\[
\langle u_n(-k_1)|u_m(-k_2)\rangle = (C_{n,k_1}^* \mathcal{T} u_n(k_1)|C_{m,k_2}^* \mathcal{T} u_m(k_2)) = C_{n,k_1} C_{m,k_2}^* \langle u_n(k_1)|u_m(k_2)\rangle^*.
\] (S6)

The velocity operator transforms as \( \mathcal{T} \hat{\nu} \mathcal{T}^{-1} = -\hat{\nu} \), and thus the velocity matrix element satisfies

\[
\langle u_n(-k_1)|\hat{\nu}|u_m(-k_2)\rangle = (C_{n,k_1}^* \mathcal{T} u_n(k_1)|\mathcal{T}(-\hat{\nu}) \mathcal{T}^{-1} C_{m,k_2}^* \mathcal{T} u_m(k_2)) = -C_{n,k_1} C_{m,k_2}^* \langle u_n(k_1)|\hat{\nu}|u_m(k_2)\rangle^*.
\] (S7)

The symmetry properties of the Bloch hamiltonian can also be constrained by other point group symmetries of the crystal. A particularly interesting example is that of mirror symmetry. For example, in the presence of mirror along the \( y \)-axis, such that \( M_y H(k) M_y^{-1} = H(M_y k) \), where \( M_y : (x, y) \rightarrow (-x, y) \). The energy dispersion thus satisfies \( \epsilon_{n,k} = \epsilon_{n,M_y k} \). On the other hand, the velocity operator transforms as \( M_y \hat{v}_z M_y^{-1} = -\hat{v}_z \) and \( M_y \hat{v}_y M_y^{-1} = \hat{v}_y \).

Following similar arguments as above, we obtain symmetry relations for wavefunctions and velocity matrix elements in much the same form as above, leading to distinctive properties of the non-vertical injection and shift photocurrents as discussed in the main text and below.

A. Symmetry analysis for non-vertical injection current

1. Inversion symmetry

When the material possesses \( \mathcal{P} \)-symmetry and identifying band indices \( n, m \) in Eq. [S4] with \( c, v \), we find that the interband velocity matrix element \( \hat{\nu}_{cv}(k, q) \) satisfies \( \hat{\nu}_{cv}(k, q) = -C_{c,k+q}^* C_{v,k-q}^* \hat{\nu}_{cv}(-k,-q) \). Thus, for linear
[denoted as $\theta$] and circularly [denoted as $\eta = \pm 1$] polarised light, the square of the transition matrix element

$$
\nu_{cv}^{\theta(\eta)}(k, q) = |E^{\theta(n)} \cdot \nu_{cv}(k, q)|^2
$$

(S8)

thus obeys $\nu_{cv}^{\theta(\eta)}(k, q) = \nu_{cv}^{\theta(\eta)}(-k, -q)$. This reproduces Eq. (8) of the main text.

Next we note when the material possesses $P$-symmetry, the group velocities in valence and conduction bands satisfy $v_c(k + q/2) = -v_c(-k - q/2)$ and $v_v(k - q/2) = -v_v(-k + q/2)$, we have $\Delta(k, q) = -\Delta(-k, -q)$ odd under $k \rightarrow -k$, $q \rightarrow -q$. On the other hand, since $\epsilon_{n,k}$ is even in $k$-space, we have $\rho(k, q) = \rho(-k, -q)$. This reproduces Eq. (7) of the main text.

Therefore, in the presence of inversion symmetry, the injection current (obtained by summing Eq. (4) of the main text over $k$-space) obeys

$$
\partial J^\text{ini}_\theta(q) = -\partial J^\text{ini}_\theta(-q), \quad \partial J^\text{ini}_\eta(q) = -\partial J^\text{ini}_\eta(-q),
$$

(S9)

as discussed in Table I of the main text.

2. Time reversal symmetry

When the material possesses $T$-symmetry, Eq. (S7) gives $\nu_{cv}(k, q) = C^c_{c,k+q/2}C^v_{v,k-q/2} \nu_{cv}(-k, -q)$. For linearly polarised light, since $E^\theta = (E^\theta)^*$, we have $v_{cv}^{\theta}(k, q) = v_{cv}^{\theta}(-k, -q)$. In contrast, for circularly polarised light, we have $v_{cv}^{\theta}(k, q) = v_{cv}^{\eta}(-k, -q)$. This latter relation can be obtained by noting $E^\eta = (E^{-\eta})^*$ for circularly polarised irradiation.

We now turn to the carrier velocity $v_c(k)$. For $T$-symmetry preserving materials, we have $v_c(k + q/2) = -v_c(-k - q/2)$ and $v_v(k - q/2) = -v_v(-k + q/2)$. Thus, the change in carrier velocity obeys $\Delta(k, q) = -\Delta(-k, -q)$. Similar to that discussed above for inversion symmetry, $T$-symmetry preserving materials also possess energy dispersion relations that are even in $k$-space yielding $\rho(k, q) = \rho(-k, -q)$.

As a result, the linear and circular injection current (obtained by summing Eq. (4) of the main text over $k$-space) obeys

$$
\partial J^\text{ini}_\theta(q) = -\partial J^\text{ini}_\theta(-q), \quad \partial J^\text{ini}_\eta(q) = \partial J^\text{ini}_\eta(-q)
$$

(S10)

as discussed in Table 1 of the main text.

Combining both Eq. (S9) and Eq. (S10), we conclude that non-vertical linear injection charge photocurrents are in general allowed in materials with both $P$- and $T$-symmetries. In contrast, non-vertical circular injection charge photocurrents (a photocurrent that depends on the helicity of the incident light) vanishes when both $P$- and $T$-symmetries in the material remain intact.

3. Mirror symmetry

It is also interesting to consider how point group symmetries can also similarly constrain the form of the non-vertical injection photocurrents. As a simple illustration we focus on non-vertical linear injection photocurrents in a material with a mirror axis along $y$. For simplicity, we consider the case where incident light (linear) polarization $|E = E_0 \hat{y}|$ as well as non-vertical transition wavevector $q$ is directed along the mirror axis $y$. As a result, for $q = q \hat{y}$, we have $M_y q = q$. In this case, the square of the transition matrix element obeys $v_{cv}^{\theta}(k, q) = v_{cv}^{\theta}(M_y k, q)$. On the other hand, the component of the change in electron group velocity normal to the mirror axis will switch sign $\Delta_y(k, q) = -\Delta_x(M_y k, q)$ while the component parallel to the mirror axis remains invariant $\Delta_y(k, q) = \Delta_y(M_y k, q)$ under mirror reflection.

We note that $\rho(k, q) = \rho(M_y k, q)$ only depends on the energy dispersion (which is even under mirror reflection). As a result, the component of the non-vertical linear injection photocurrent normal $q$ (when it is directed along the mirror axis) vanishes: $[\partial J^\text{ini}_\theta]_y(q) = 0$, while the component parallel to $q$ when it is directed along the mirror axis, $[\partial J^\text{ini}_\theta]_y(q)$, is allowed.
B. Symmetry analysis for non-vertical shift photocurrent

To investigate the symmetry properties of the non-vertical shift photocurrent, it is useful to re-write the shift vector \( \mathbf{r} \) in terms of a derivative of the phase of the Wilson loop associated with the interband transition \([20][21]\):

\[
\mathbf{r}(\mathbf{k}, \mathbf{q}) = \lim_{\delta \mathbf{p} \to 0} \nabla_{\delta \mathbf{p}} \arg \mathcal{W}(\mathbf{k}, \delta \mathbf{p}, \mathbf{q}),
\]

where the Wilson loop is defined as

\[
\mathcal{W}(\mathbf{k}, \delta \mathbf{p}, \mathbf{q}) = \langle \psi_{\nu}(\mathbf{k} - \mathbf{q}/2) | \psi_{\nu}(\mathbf{k} + \delta \mathbf{p} - \mathbf{q}/2) | \hat{\mathbf{e}} \cdot (\langle \psi_{\nu}(\mathbf{k} + \delta \mathbf{p} - \mathbf{q}/2) | \hat{\mathbf{e}} | \psi_{\nu}(\mathbf{k} + \delta \mathbf{p} + \mathbf{q}/2) \rangle \rangle \langle \psi_{\nu}(\mathbf{k} + \mathbf{q}/2) | \psi_{\nu}(\mathbf{k} + \mathbf{q}/2) \rangle \langle \psi_{\nu}(\mathbf{k} - \mathbf{q}/2) | \psi_{\nu}(\mathbf{k} - \mathbf{q}/2) \rangle,
\]

where \( \mathbf{e} \) is the unit vector indicating the electric field polarisation direction. For linearly polarised light, we have \( \hat{\mathbf{e}} = \hat{n} = \hat{x} + i \hat{y} \). For circularly polarised light, we have \( \hat{\mathbf{e}} = \hat{e}_\theta = \hat{x} \cos \theta + \hat{y} \sin \theta \).

In the following, we analyse the symmetry properties of the non-vertical shift photocurrent by examining the Wilson loop \( \mathcal{W}(\mathbf{k}, \delta \mathbf{p}, \mathbf{q}) \) under various symmetries.

1. Inversion symmetry

When the material possesses \( \mathcal{P} \)-symmetry, the inner product of the wavefunctions follows the relation in Eq. (S3) while the velocity matrix element obeys Eq. (S4). Since all the Bloch wavefunctions in Eq. (S12) occur in pairs (guaranteeing its gauge invariance), the phase factors for the wavefunctions resulting from the \( \mathcal{P} \) transformation fully compensate with each other. As a result, we find

\[
\mathcal{W}(\mathbf{k}, \delta \mathbf{p}, \mathbf{q}) = -\mathcal{W}(-\mathbf{k}, -\delta \mathbf{p}, -\mathbf{q}), \quad \arg \mathcal{W}(\mathbf{k}, \delta \mathbf{p}, \mathbf{q}) = \arg \mathcal{W}(-\mathbf{k}, -\delta \mathbf{p}, -\mathbf{q}) + \pi.
\]

Since the shift vector \( \mathbf{r}(\mathbf{k}, \mathbf{q}) \) depends on the derivative of \( \arg \mathcal{W}(\mathbf{k}, \delta \mathbf{p}, \mathbf{q}) \), we arrive at

\[
\mathbf{r}(\mathbf{k}, \mathbf{q})^{\theta(\eta)} = -\mathbf{r}^{\theta(\eta)}(-\mathbf{k}, -\mathbf{q}),
\]

reproducing Eq. (9) of the main text.

To understand the symmetry properties of the non-vertical shift charge photocurrent, we recalling that both \( \nu^\eta(\mathbf{k}, \mathbf{q}) \) and \( \rho(\mathbf{k}, \mathbf{q}) \) are even under \( \mathcal{P} \) symmetry. By summing Eq. (5) of the main text over \( k \)-space, we find that the non-vertical shift photocurrents flow in opposite directions for \( \pm \mathbf{q} \) in \( \mathcal{P} \)-preserving materials:

\[
\mathbf{j}^\text{shift}_\theta (\mathbf{q}) = -\mathbf{j}^\text{shift}_\theta (-\mathbf{q}), \quad \mathbf{j}^\text{shift}_\text{cir} (\mathbf{q}) = -\mathbf{j}^\text{shift}_\text{cir} (-\mathbf{q}).
\]

as shown in Table I of the main text.

2. Time reversal symmetry

Following similar analysis for the injection current, in \( \mathcal{T} \)-symmetry preserving materials, we have \( \hat{\mathbf{e}}_\theta \cdot \nu_{\text{cir}}(\mathbf{k}, \mathbf{q}) = -C'_{\nu, \mathbf{k} + \mathbf{q}/2} C'_{\nu, \mathbf{k} - \mathbf{q}/2} [\hat{\mathbf{e}}_\theta \cdot \nu_{\text{cir}}(-\mathbf{k}, -\mathbf{q})]^* \) for linearly polarised light. Combining with the relation for the Bloch wavefunctions in Eq. (S6), we obtain

\[
\mathcal{W}^\theta(\mathbf{k}, \delta \mathbf{p}, \mathbf{q}) = -[W^\theta(-\mathbf{k}, -\delta \mathbf{p}, -\mathbf{q})]^*, \quad \arg \mathcal{W}^\theta(\mathbf{k}, \delta \mathbf{p}, \mathbf{q}) = -\arg \mathcal{W}^\theta(-\mathbf{k}, -\delta \mathbf{p}, -\mathbf{q}) + \pi.
\]

Here we note that the additional phase factors \( C' \) that arise under \( \mathcal{T} \) transformation fully compensate each other since the Bloch wavefunctions in Eq. (S12) occur in pairs.

By taking the derivative of the phase of \( \mathcal{W}^\theta(\mathbf{k}, \delta \mathbf{p}, \mathbf{q}) \), we arrive at the symmetry constraint for the shift vector (for linearly polarised light) in \( \mathcal{T} \)-preserving materials:

\[
\mathbf{r}^{\theta}(\mathbf{k}, \mathbf{q}) = -\mathbf{r}^{\theta}(-\mathbf{k}, -\mathbf{q}).
\]

as discussed in Eq. (9) of the main text.
On the other hand, for circularly polarised light, we have $\hat{e}^\eta \cdot \nu_{cv}(k, q) = -C^\eta_{c,k+q/2}C^{*\eta}_{c,k-q/2} [\hat{e}^{-\eta} \cdot \nu_{cv}(-k, -q)]^\dagger$. Similarly, the Wilson loop satisfies

$$W^\eta(k, \delta p, q) = -[W^{-\eta}(-k, -\delta p, q)]^\dagger, \quad \arg[W^\eta(k, \delta p, q)] = -\arg[W^{-\eta}(-k, -\delta p, -q)] + \pi. \quad (S18)$$

As a result, we find that the shift vector (for circularly polarized light with helicity $\eta$) satisfies

$$r^\eta_0(k, q) = r^{-\eta}(-k, -q). \quad (S19)$$

as discussed in Eq. (9) of the main text.

By summing Eq. (6) of the main text over $k$-space, the non-vertical linear and circular shift charge photocurrents in $T$-symmetric materials obey

$$j^\text{shift}_\eta(q) = j^\text{shift}_\eta(-q), \quad j^\text{shift}_{c\eta}(q) = -j^\text{shift}_{c\eta}(-q). \quad (S20)$$

Combining with the constraints in Eq. (S15) and (S20), we find that in $P$- and $T$-symmetric materials, non-vertical linear shift charge photocurrent vanishes for all non-vertical wavevectors $q$ while non-vertical circular shift charge photocurrent are allowed.

3. Mirror symmetry

Here we illustrate how mirror symmetry can constrain the form of the non-vertical circular shift photocurrent. As a simple illustration we focus on a mirror plane axis along $y$ and consider non-vertical transition wavevector $q$ directed along the mirror axis $y$.

For circularly polarised light with polarisation vector $\hat{e}^\eta$, the square of the transition matrix element satisfies $\nu_{cv}^\eta(k, q) = \nu_{cv}^{\eta\dagger}(M_y k, q)$. The Wilson loop obeys

$$W^\eta(k, \delta p, q) = -W^{-\eta}(M_y k, M_y \delta p, q), \quad \arg[W^\eta(k, \delta p, q)] = \arg[W^{-\eta}(M_y k, M_y \delta p, q)] + \pi. \quad (S21)$$

By taking the derivative with respect to $\delta p$, we have

$$r^\eta_y(k, q) = -r^{-\eta}_x(M_y k, q), \quad r^\eta_y(k, q) = r^{-\eta}_x(M_y k, q). \quad (S22)$$

Since $\rho(k, q) = \rho(M_y k, q)$, the $y$-component of the helicity dependent charge circular shift current vanishes while $[j^\text{shift}_{c\eta}]_x$ is allowed, i.e. non-vertical charge circular shift current in the presence of $M_y$-symmetry is purely transverse.

II. $c, v$ band symmetry and non-vertical photocurrents in bilayer graphene

In this section, we discuss how a symmetry between the conduction $c$ and valence $v$ bands can emerge in the low-energy effective Hamiltonian for bilayer graphene. As we will show below, this effective $c, v$ band symmetry leads to a vanishing non-vertical linear injection and circular shift photocurrents at low temperature when the Fermi surface (determined by $\mu$) does not intersect and are far from the interband transition contours (determined by $h\omega$).

We consider the low energy Hamiltonian in Eq. (10) in the main text. For a two-band Hamiltonian, we can directly solve for the eigenenergies and eigenstates. As discussed above, these enable to directly compute the shift vector $r^\eta_0(p, q)$ and the change in carrier velocity $\Delta\nu(p, q)$ as a carrier is photoexcited between $c$ and $v$ bands. For the convenience of the reader, we rewrite $H(p)$ as

$$H(p) = d_1 \sigma_x + d_2 \sigma_y, \quad d_1 = -\frac{\hbar^2}{2m} (p_x^2 - p_y^2) + hv_3 \zeta p_x, \quad d_2 = -\frac{\hbar^2}{m} \zeta p_x p_y - hv_3 p_y. \quad (S23)$$

The energy dispersion is given by $\epsilon_{c,v}(p) = \pm \sqrt{d_1^2 + d_2^2}$, where the explicit $\zeta$ and $p$ dependence of $d_1$ and $d_2$ is suppressed for brevity. The corresponding eigenstates are

$$|u_\zeta^c(p)\rangle = \frac{1}{\sqrt{2}} \begin{pmatrix} e^{-i\zeta\phi_c} \\ 1 \end{pmatrix}, \quad |u_\zeta^v(p)\rangle = \frac{1}{\sqrt{2}} \begin{pmatrix} e^{-i\zeta\phi_v} \\ -1 \end{pmatrix}, \quad (S24)$$

where $\phi_{c,v} = \tan^{-1} (d_2/d_1)$. As we now show, for $P$- and $T$-symmetric bilayer graphene, there is an additional emergent symmetry between the conduction and the valence bands that relates the conduction and valence bands in the separate valleys.
To see this, first we note that (as can be verified by inspection) the energies of the conduction and valence bands in the separate valleys obey \( \epsilon_{c}^{\pm}(p) = -\epsilon_{v}^{\pm}(p) \). This yields a non-vertical interband transition energy:

\[
\epsilon_{cv}^{\pm}(p, q) = \epsilon_{c}^{\pm}(p + q/2) - \epsilon_{v}^{\pm}(p - q/2) \quad \text{that obeys}
\]

\[
\epsilon_{cv}^{\pm}(p, q) = \epsilon_{cv}^{\pm}(-p, q)
\] (S25)

This relationship between \( c \) and \( v \) bands between valleys also constrains the interband velocity matrix elements. Using Eq. (S24), we can explicitly compute \( \nu_{cv}^{\pm}(p, q) = \langle u_{c}^{\pm}(p + q/2)|\hat{\nu}|u_{v}^{\pm}(p - q/2) \rangle \) [where \( \hat{\nu} = \nabla_{p} H(p)/\hbar \)] as

\[
\nu_{cv,x}^{\pm}(p, q) = \frac{1}{2} \left( -\frac{h}{m} p_{x} + v_{3} \right) \left[ e^{i\phi_{c,p+q/2} - e^{i\phi_{v,p-q/2}}} - i \frac{h}{2m} \zeta p_{y} \right] \left[ e^{i\phi_{c,p+q/2} + e^{i\phi_{v,p-q/2}}} \right]
\]

\[
\nu_{cv,y}^{\pm}(p, q) = \frac{1}{2} \frac{h}{m} p_{y} \left[ -e^{i\phi_{c,p+q/2} + e^{i\phi_{v,p-q/2}}} - i \frac{h}{2m} \zeta p_{x} + v_{3} \right] \left[ e^{i\phi_{c,p+q/2} + e^{-i\phi_{v,p-q/2}}} \right]
\] (S26)

We note that when \( \zeta \to -\zeta \), \( p \to -p \), we have \( d_{1} \to d_{1} \) and \( d_{2} \to -d_{2} \) [see Eq. (S23)]. Thus, \( \phi_{\zeta, p} = -\phi -\zeta, -p \) is odd in \( k \)-space, and we have

\[
\nu_{cv}^{\pm}(p, q) = \nu_{cv}^{\pm}(-p, q).
\] (S27)

As a result, the square of the interband transition matrix for linearly (circularly) polarised light obeys \( \nu_{\zeta,\theta}(p, q) = \nu_{-\zeta,\theta}(-p, q) \).

The above symmetry relations for how velocity matrix element (and the energies) transform as \( \zeta \to -\zeta \), \( p \to -p \) can be directly used to determine the the non-vertical circular shift photocurrent. To proceed, we consider the shift vector for circularly polarized light reproduced here for the convenience of the reader as

\[
r_{\zeta,\eta}^{\pm}(p, q) = [A_{c}^{\pm}(p + q/2) - A_{v}^{\pm}(p - q/2)] - \nabla_{p} \arg (E^{\eta} \cdot \nu_{cv}^{\pm}(p, q))
\] (S28)

By direct computation using Eq. (S24), we find the Berry connection in the valence and conduction bands in opposite valleys satisfy \( A_{c}^{\pm}(p) = A_{v}^{\pm}(-p) \). This means that the difference of Berry connections (square brackets in Eq. (S28)) is odd when \( \zeta \to -\zeta \), \( p \to -p \), namely: \( A_{c}^{\pm}(p + q/2) - A_{v}^{\pm}(p - q/2) = -[A_{c}^{-\zeta}(-p + q/2) - A_{v}^{-\zeta}(-p - q/2)] \). Further, by applying Eq. (S27) to the last term of Eq. (S28) we find: \( \nabla_{p} \arg (E^{\eta} \cdot \nu_{cv}^{\pm}(p, q)) \) is also odd as \( \zeta \to -\zeta \), \( p \to -p \). Hence, we find that as \( \zeta \to -\zeta \), \( p \to -p \) the weighted shift vector \( R_{\zeta,\eta}^{\pm}(p, q) \equiv \nu_{\zeta,\theta}^{\pm}(p, q)r_{\zeta,\eta}^{\pm}(p, q) \) obeys

\[
R_{\zeta,\eta}^{\pm}(p, q) = -R_{-\zeta,\eta}^{\pm}(-p, q)
\] (S29)

This is verified in Fig. 2(b) and (c) in the main text, which shows the numerical vector plot for \( R_{\zeta,\eta}^{\pm}(p, q) \). Finally, we note that when the Fermi surface is far from any interband transition contours such that \( f_{cv}^{\pm}(p, q) \) is a constant for all \( p \), we have

\[
\rho_{\zeta}^{\pm}(p, q) = \rho_{-\zeta}^{\pm}(-p, q)
\] (S30)

This can be achieved, for instance, for a large \( h\omega \) and chemical potential fixed close to charge neutrality at low temperature. In this case, by summing the expression for the non-vertical circular shift photocurrent in Eq. (S5) of the main text across all \( p \) and both valleys, we find the non-vertical circular shift photocurrent vanishes due to the emergent symmetry between the valence and the conduction bands.

A similar argument can also be applied to the non-vertical linear injection photocurrent. The change in electron group velocity \( \Delta \zeta^{\pm}(p, q) \) can be written as \( \Delta \zeta^{\pm}(p, q) = \nabla_{p} \epsilon_{cv}^{\pm}(p, q)/\hbar \). Since \( \epsilon_{cv}^{\pm}(p, q) = \epsilon_{cv}^{\pm}(-p, q) \), we have \( \Delta \zeta^{\pm}(p, q) = -\Delta \zeta^{\pm}(-p, q) \). In the same fashion as discussed above, when the Fermi surface is far from any interband transition contours such that \( f_{cv}^{\pm}(p, q) \) is a constant for all \( p \), we have Eq. (S30). As a result, in such a situation, applying Eq. (S27), (S30), as well as \( \Delta \zeta^{\pm}(p, q) = -\Delta \zeta^{\pm}(-p, q) \), and summing the expression for the non-vertical linear injection photocurrent in Eq. (4) of the main text across all \( p \) and both valleys, we find a vanishing non-vertical linear injection current.

Of course, a finite non-vertical circular shift photocurrent as well as non-vertical linear injection photocurrent can be produced when either \( f_{cv}^{\pm}(p, q) \) is not constant for all \( p \) (this is the Fermi surface resonant case discussed in the main text), or when the emergent \( c, v \) band symmetry is broken.