Visualization of bulk and edge photocurrent flow in anisotropic Weyl semimetals

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Materials that rectify light into current in their bulk are desired for optoelectronic applications. In Weyl semimetals that break inversion symmetry, bulk photocurrents may arise due to nonlinear optical processes that are enhanced near the Weyl nodes. However, the photoresponse of these materials is commonly studied by scanning photocurrent microscopy, which convolves the effects of photocurrent generation and collection. Here we directly image the photocurrent flow inside the type-II Weyl semimetals WTe\(_2\) and TaIrTe\(_4\) using high-sensitivity quantum magnetometry with nitrogen-vacancy centre spins. We elucidate a mechanism for bulk photocurrent generation, which we call the anisotropic photothermoelectric effect, where unequal thermopowers along different crystal axes drive intricate circulations of photocurrent around the photoexcitation. Using overlapping scanning photocurrent microscopy and magnetic imaging at the interior and edges of the sample, we visualize how the anisotropic photothermoelectric effect stimulates the long-range photocurrent collected in our WTe\(_2\) and TaIrTe\(_4\) devices through the Shockley–Ramo mechanism. Our results highlight a widely relevant source of current flow and will inspire photodetectors that utilize bulk materials with thermoelectric anisotropy.

For directional photocurrent flow in the absence of a bias voltage, symmetry breaking is an essential ingredient\(^1\)–\(^3\). In devices, symmetry is commonly broken by joining dissimilar materials or different dopings of the same material, driving photocurrent flow through a difference in their Seebeck coefficients\(^6,7\) or through the built-in electric field at a p–n junction\(^8,9\). Generating photocurrents throughout a single homogeneous material can, however, be advantageous. One approach utilizes the temporal asymmetry in the exciting light field, injecting currents from the interference of coherent electron dynamics driven by ultrafast pulses\(^10,11\). Alternatively, the bulk photovoltaic effect (BPVE), a nonlinear optical process exhibited by non-centrosymmetric crystals\(^12–14\), generates a directional photocurrent from the asymmetry in the electron wavefunctions before and after photoexcitation.

Non-magnetic Weyl semimetals, which feature topological band touchings preserved by the breaking of inversion symmetry, are compelling candidates to host the BPVE. Recent insight has cast the BPVE in terms of the quantum geometric properties of the band structure, predicting an enhanced response for low-energy excitation near the Weyl nodes\(^14–16\). Indeed, experiments on non-centrosymmetric Weyl semimetals, including WTe\(_2\) (ref. \(^17\)), TaIrTe\(_4\) (refs. \(^18,19\)), MoTe\(_2\) (ref. \(^18\)) and TaAs (refs. \(^20,21\)), have demonstrated signatures of shift and injection currents that correspond to the components of BPVE controlled by linearly and circularly polarized light, respectively. However, these observations rely on scanning photocurrent microscopy (SPCM)\(^17–22\) or terahertz emission\(^23,24\), neither of which can resolve the microscopic details of the photocurrent flow. In SPCM, a focused laser beam is...
rastered on a device and the total current between two distant contacts is recorded. For gapless materials, the global diffusive photocurrent collected by SPCM is indirectly induced by the intrinsic photocurrent local to the photoexcitation through the Shockley–Ramo mechanism. This local photocurrent is invisible to SPCM, yet captures the essential light–matter interaction.

To clarify their photocurrent response, here we directly visualize the two-dimensional (2D) photocurrent flow in the type-II Weyl semimetals WTe$_2$ and TaIrTe$_4$. Our unique technique, which we call photocurrent flow microscopy (PCFM), is enabled by the high-sensitivity, high-spatial-resolution magnetometry of the photocurrent’s Oersted field through the coupled spin precession of nitrogen-vacancy (NV) centres (515 nm). Subsequent relaxation leads to heating for the Weyl semimetal’s crystal lattice. This local flow pattern, explained by our theoretical and ab initio simulations, reveals that the primary photocurrent mechanism in our devices is unrelated to nonlinear optical processes or Weyl excitations. Rather, it originates from a robust but overlooked broken symmetry of the bulk: anisotropy in the in-plane transport. Using coincident SPCM and PCFM imaging, we visualize how this anisotropic photothermoelectric effect (APTE) stimulates the collected photocurrent through the Shockley–Ramo framework when exciting low-symmetry edges and in the device interior for particular contact geometries. Our observations open concepts for broadband, position-sensitive photodetectors using homogeneous materials with intrinsic Seebeck anisotropy.

Figure 1a displays our room-temperature experimental configuration (Supplementary Section 1) that extends another work that pioneered the detection but not the 2D imaging of photocurrents with NV ensemble magnetometry. Here we use a thin diamond membrane to optically access non-transparent photocurrent samples and improve the NV photon collection efficiency for the challenging imaging measurements required for the model-free reconstruction of photocurrent flow. By transferring exfoliated flakes onto prefabricated electrodes on the diamond substrate, we are able to perform both PCFM and SPCM in situ on the same device. In PCFM, the NV-centre probe beam (green, 515 nm) is scanned around a fixed position for the photocurrent excitation beam (red, 661 nm) to map the local magnetic field. Alternatively, by measuring the total collected current as either beam is rastered, we can acquire SPCM images at either wavelength.

Type-II Weyl semimetals WTe$_2$ and TaIrTe$_4$ crystallize in an inversion-breaking orthorhombic structure (space group $Pmn_2_1$). As shown in Fig. 1b, for a single layer of Td-WTe$_2$, the transition metal atoms form quasi-one-dimensional zigzag chains along the $a$ axis within the $a$–$b$ plane, leading to anisotropic electrical, optical and thermal properties. The lowest-order shift photocurrent is described by $J_i = \Re (\sigma^{(2)}_{gk}E_i(\omega)E^*_k(\omega))$ where $\Re (\sigma^{(2)}_{gk})$ is the real part of the second-order susceptibility tensor and $E(\omega) = E_0 e^{i\omega t}$ are the Cartesian components.
of the light’s electric field\textsuperscript{12–15}. For a system with inversion symmetry, $\sigma_{i\ell}$ must identically vanish since $E_{\ell}(\omega)E_{i}^{*}(\omega)$ is even under inversion, whereas $I_{i\ell}$ is odd. Moreover, for light at normal incidence, a shift current in the $a$–$b$ plane of WTe\textsubscript{2} or TaI\textsubscript{2}Se\textsubscript{3} is prohibited by a similar argument using the two-fold rotation symmetry ($C_2$ screw axis) within the $a$–$b$ plane (Fig. 1B)\textsuperscript{18–20}.

In SPCM experiments on WTe\textsubscript{2} (ref. \textsuperscript{17}) and TaI\textsubscript{2}Se\textsubscript{3} (refs. \textsuperscript{18,20}), photocurrents in the $a$–$b$ plane were detected when exciting mirror-symmetry-breaking edges and within the sample’s bulk near narrow electrical contacts. To justify the nominally forbidden interior photocurrent, another work\textsuperscript{20} interpreted the bulk response in TaI\textsubscript{2}Se\textsubscript{3} as a third-order process $J_i = \sigma_{i\ell}^{(3)}E_{\ell}E_{\ell}^{*}E_{\ell}^{(w)} (\omega)$, where the normally incident light field mixes with an uncontrolled d.c. electric field $E_{\ell}^{(w)}$ that arises from device-specific interfaces or inhomogeneities. Alternatively, in the near-field SPCM experiment\textsuperscript{18}, it was argued that the bulk signal arises from non-planar second-order susceptibility elements (for example, $\sigma_{m\ell}$ and $\sigma_{mn\ell}$) that become accessible due to an out-of-plane electric field $E_{\ell}^{(w)} (\omega)$ created by the metallic tip. Although such mechanisms may contribute, we reveal that the uncovered APTE is a unified cause of bulk and edge photocurrents in WTe\textsubscript{2} and TaI\textsubscript{2}Se\textsubscript{3}.

To achieve the enhanced sensitivity to detect photocurrent densities lower than microamperes per micrometre, we utilize a multipulse quantum lock-in sequence for a.c. magnetic fields\textsuperscript{29,33–35}. As shown in Fig. 1c, we strobe the photocurrent excitation on the sample and simultaneously apply a sequence of microwave π-pulses (XY8-quantum lock-in sequence for a.c. magnetic fields\textsuperscript{29,33–35}). As shown in Fig. 1d, the evolution of the NV superposition state $|\psi\rangle$ on the Bloch sphere at various times in a measurement sequence with timing delay $\theta \approx 0.11\pi \text{ rad}$ between the photocurrent and microwave pulses. The final state is rotated by a π/2-pulse around either the $x$ or $y$ axis of the Bloch sphere and then read by the NV fluorescence to yield the projections $X_\psi$ or $Y_\psi$ from which the state’s precession angle $\Delta\varphi$ is determined by $\Delta\varphi = \arctan(Y_\psi/X_\psi)$ (Methods).

We first demonstrate photocurrent detection for linearly polarized light in the interior of a WTe\textsubscript{2} sample (device A). Figure 2a measures the NV spin precession versus delay $\theta$ for a fixed spatial offset $3 \, \mu\text{m}$ vertically) between the photocurrent and NV probe beams.

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**Fig. 2 | Crystal-axes-aligned bulk photocurrents in WTe\textsubscript{2}.** a, Projections $X_\psi$ and $Y_\psi$ of the final NV state versus delay $\theta$ between the photocurrent and NV spin-driving pulses. The error bars in the data represent one standard deviation in the photon shot noise. The probed NV position is the green circle in d. We extract the maximum NV phase precession $\Delta\varphi = 1.30 \pm 0.07 \text{ rad}$, corresponding to a magnetic field $B_{pc} = 140 \pm 8 \text{ nT}$ along the NV centre axis, where the error bar denotes the 95% confidence interval on the fitted parameter, for a photocurrent excitation (red) power $P = 120 \mu\text{W}$. b, Precession angle $\Delta\varphi$ versus optical power $P$. The linear dependence of $\Delta\varphi$ on $P$ is consistent with electrical measurements of the global photocurrent $I_{ph}$ between the photocurrent and NV fluorescence at the centre of the image ($P = 120 \mu\text{W}$). c, Optical micrograph of WTe\textsubscript{2} device A with a sample thickness of 1.8 $\mu\text{m}$. The long edges identify the $a$ axis of WTe\textsubscript{2}. The in-plane projection of $B_{pc} (28 \text{ mT})$ is denoted by the white arrow, but has no influence on the results. d, Spatial image of the NV centre precession angle $\Delta\varphi (r) = B_{pc}(r)$ in the interior of WTe\textsubscript{2} device A, with the photoexcitation fixed at the centre of the image ($P = 120 \mu\text{W}$). The imaged region is the red box in c. e, Photocurrent density $J(r)$ reconstructed from $B_{pc}(r)$ in d as a 2D vector field. The radial component of the flow $(J \cdot r)$ is superimposed as a false-colour map. The local photocurrent circulates by flowing in along the $a$ axis and flowing out along the $b$ axis. f, Optical micrograph of WTe\textsubscript{2} device B, with a sample thickness of 360 nm. g, Spatial image of $B_{pc}(r)$ for the boxed region in f with $P = 120 \mu\text{W}$. h, Reconstructed $J(r)$ in device B. The fourfold photocurrent circulation remains aligned with the crystal axes of the rotated flake.
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Thus, our imaging (Fig. 2e,h) identifies the \(a\) axis as possessing the more negative Seebeck coefficient \(S_a < S_b\). Assuming \(|S_a - S_b| = 10 \mu V K^{-1}\) and an approximately isotropic conductivity \(\sigma_a/\sigma_b = 1\) (Supplementary Fig. 4), the equilibrium potential \(-\Phi(r)\) corresponding to \(\Delta \psi(r)\) can be obtained (Fig. 3b). Since the electromotive force on electrons points towards smaller values (blue) of \(-\Phi(r)\), Fig. 3b illustrates how \(\Phi\) drives electron backflow opposite to \(\nabla \psi\), as well as electron circulation from the \(a\) axis to the \(b\) axis. In Fig. 3c, we calculate the full photocurrent distribution \(J(r)\) using equation (1), showing excellent agreement with our WTe\(_2\) images (Fig. 2e,h). A quantitative estimate of Seebeck anisotropy is complicated by the dependence of \(J\) on not only \(|S_a - S_b|\) but also on conductivities \(\sigma_{an}\) and temperature \(\Delta T\) of the WTe\(_2\) flake that cannot be directly measured. Estimating these factors, we describe our best experimental determination as \(|S_a - S_b| = 8.3^\pm 0.6 \mu V K^{-1}\) (Supplementary Section 5).

Notably, our density functional theory calculations also predict negative Seebeck coefficients \(S_a\) and \(S_b\) for WTe\(_2\) at room temperature, with \(S_a < S_b\) in agreement with experiment (Methods). For semimetals, both electron and hole bands contribute to the thermopower, with opposite signs and weighting factor determined by the band's density of states and squared group velocity in the direction of transport. As shown in Extended Data Fig. 3d, the predicted \(S_a\) and \(S_b\) values diverge above 200 K, with \(S_a\) becoming more negative and \(S_b\) becoming more positive, leading to \(|S_a - S_b| = 15 \mu V K^{-1}\) at 300 K. This trend is mainly contributed by the anisotropy in electron velocities for low-lying conduction bands. As temperature increases, the thermopower senses the larger density of states for holes away from the Fermi level, but this positive contribution is outweighed by the electron contribution along the \(a\) axis, where the electron velocities are the highest.

**Fig. 4 | APTE as the stimulus for long-range photocurrent in WTe\(_2\) devices.**

a. Experimental SPCM image of \(i_{\text{global}}\) in WTe\(_2\) device B for \(P = 300 \mu W\) at 661 nm. Robust edge photocurrents are detected along the \((110)\) and \((1\overline{1}0)\) edges, with a negative (blue) and positive (red) polarity, respectively. Sign-switching interior photocurrents are also collected relative to two localized points on the upper and lower gold pads (orange). b. Coincident PCFM image of \(J(r)\) for photoexciting the \((1\overline{1}0)\) edge in device B. c. PCFM image for photoexciting the \((1\overline{1}0)\) edge. The photoexcitation power in b and c is 120 \(\mu W\), and the locations are labelled in a. The polarity of edge photocurrent \(i_{\text{local}}\) is visualized by the net component of \(J(r)\) parallel to the edge. d. PCFM image for photoexciting the \((100)\) edge in WTe\(_2\) device A with \(P = 100 \mu W\). e - g. Simulated \(J(r)\) for the \((1\overline{1}0)\) (e), \((1\overline{1}0)\) (f) and \((100)\) (g) edges, respectively, using the APTE model. h. Simulation of the full SPCM image under the Shockley–Ramo theory. The simulation assumes that \(\Omega(r)\) flows according to anisotropic Seebeck coefficients and that the weighting field \(\nabla \psi(r)\) is determined by the boundary conditions \(\psi = 1, 0\) on two point-like surfaces labelled on the upper and lower pads, respectively.
To establish the generality of APTE, we perform PCFM imaging on TaIrTe₄, where the a-axis zigzag chains are composed of alternating Ta and Ir atoms. TaIrTe₄ is expected to exhibit more anisotropic in-plane electrical transport than WTe₂, with higher conductivity along the a axis. Our PCFM image of TaIrTe₄ shows that photoexcited electrons flow out along the a axis and flow in along the b axis (Fig. 3d), indicating Sₓ < Sᵧ (similar to WTe₂). However, a stronger asymmetry is observed, where the flow remains parallel to the a axis longer than it does to the b axis (Extended Data Fig. 4b shows the corresponding NV phase image). This distinct shape for \( \mathbf{J}(r) \) is reproduced in our APTE model (Fig. 3e) by using a higher conductivity anisotropy \( \sigma_{a/b} = 4 \) (Supplementary Section 4)\(^{48}\).

We now connect the APTE to the global photocurrent collected in our Weyl semimetal devices. In conducting materials, photocurrent collection can be mapped onto the Shockley–Ramo theorem if the timescale for charge dynamics is substantially faster than photocurrent generation\(^{35,46}\). This analogy provides the insight that the charge flowing into a distant contact \( I_{\text{global}} \) is dominated by the induced diffusion current \( I_{\text{ph}} \), rather than by the directly photoexcited current \( I_{\text{ph}} \) that remains local to the illumination. Another work\(^{48}\) showed that \( I_{\text{global}} \) can be simply calculated as

\[
I_{\text{global}} = C \int \mathbf{J}_{\text{ph}}(x,y) \cdot \nabla \Psi(x,y) \, dx \, dy
\]

using only the direct photocurrent \( J_{\text{ph}}(x,y) \) due to the anisotropic Seebeck flow and an auxiliary weighting potential \( \Psi(x,y) \) determined by solving Laplace's equation within the particular device geometry (Supplementary Section 7). The constant \( C \) depends on circuit resistances. Thus, non-zero \( I_{\text{global}} \) can generally arise if the areas of \( J_{\text{ph}} \cdot \nabla \Psi \) to the integral do not cancel due to differing symmetries for \( \mathbf{J}_{\text{ph}} \) and \( \nabla \Psi \).

Figure 4a displays our SPCM image at 661 nm excitation for WTe₂ device B (Fig. 2f). Long-range photocurrents are detected along the entirety of the oblique (110) and (1\|0) edge, with a negative and positive polarity, respectively. In addition, we collect \( I_{\text{global}} \) in the interior of the device, with a sign-switching pattern surrounding two hot spots along the upper and lower contacts. In contrast, no global photocurrent is collected by SPCM along a high-symmetry (100) edge, as shown for a separate TaIrTe₄ device (Extended Data Fig. 5). These spatial patterns reproduce the salient features, attributed to nonlinear responses, in prior SPCM measurements\(^{15-17}\).

To reveal the microscopic stimulus for our global photoresponse, we switch to PCFM imaging on WTe₂ device B with the photocurrent laser fixed at \( I_{\text{global}} \) hot spots. Figure 4b,c displays the photocurrent flow when exciting the upper (1\|0) and lower (110) edges, respectively. The local photocurrent distribution appears as though the edge truncates the fourfold APTE pattern observed in the interior of the WTe₂ device (Fig. 2e,h). This is corroborated in Fig. 4d, where we image the photocurrent flow when exciting a (100) edge on WTe₂ device A (Extended Data Fig. 6). The visual interpretation of these images motivates the presence or absence, as well as polarity, of global edge photocurrents. For photoresponsive oblique edges (for example, (100) and (1\|0); Fig. 4b,c, respectively), \( \mathbf{J}(r) \) possesses a strong component parallel to the edge, with the direction set by the truncation of the APTE flow inwards along the a axis and outwards along the b axis. For non-responsive (100) or (010) edges (Fig. 4d), the component of \( \mathbf{J}(r) \) parallel to the edge cancels out, consistent with the mirror symmetry about the perpendicular to these edges.

More precisely, NV magnetometry senses the total photocurrent \( \mathbf{J}(r) \) contributed by both Seebeck current \( J_{\text{ph}} \) and diffusion current \( J_{\text{d}} \). For APTE, \( \mathbf{J}_{\text{ph}} \) possesses both strong local (circulating) component and weaker long-range component, where the latter can be extracted by the contact. Our measured \( I_{\text{global}} \), mainly reflecting the long-range component of \( J_{\text{ph}} \), is only a few nanoamperes. Meanwhile, the total current density \( \mathbf{J}(r) \) near the edge is of the order of 100 nA μm⁻¹, indicating that the source/drain effect of the contacts is a small perturbation. Hence, we compare our data (Fig. 4b–d) with local simulations of \( \mathbf{J}(r) \) due to APTE (equation (1)) without considering the distant contacts (Supplementary Fig. 9 shows the simulations over the whole device geometry that include equipotential conditions at the contacts). Figure 4e–g presents the simulated APTE patterns that incorporate the modified temperature distribution and parallel-flow boundary condition near the edge for the (110), (1\|0) and (100) edges, respectively, showing excellent agreement with experiment (Fig. 4b–d).

Continuing, we simulate the full SPCM image via the Shockley–Ramo theorem (equation (2)). We approximate \( I_{\text{global}}(r,r_s) \) for each photocollection location \( r_s \) by assuming that the temperature distribution \( T(r,r_s) \) is cropped by the device boundaries if \( r_s \) is proximal to the edge. The details of \( I_{\text{global}}(r,r_s) \) also depend on \( \nabla \psi(r) \), which is determined by how electrical contact is made to the device. In WTe₂ device B, our modelling (Fig. 4h) reveals that the experimental \( I_{\text{global}} \) image can be reproduced by a simple point-like contact (white circle) in each of the upper and lower leads. Evidently, although our bottom contacts appear extended, they form effective point-like conductive interfaces in this device. We confirm that \( \mathbf{J}(r) \) near the contacts (Extended Data Fig. 1e) remains virtually identical to that in the interior of the device (Fig. 2h).

Thus, although \( J_{\text{ph}}(r) \) is \( C \), symmetric in the bulk, non-zero \( I_{\text{global}} \) can still be collected for non-uniform \( \nabla \psi \). Our bulk photocurrent, thus, stems from the interaction between \( J_{\text{ph}}(r) \) and the changing direction and strength of \( \nabla \psi \) as the point-like contacts are approached (Extended Data Fig. 1c and Supplementary Fig. 10). We observe qualitatively identical long-range edge and interior photocurrents in TaIrTe₄ devices (Extended Data Figs. 4 and 5), establishing APTE as a ubiquitous and primary mechanism in anisotropic Weyl materials.

Conclusions and outlook: we have introduced a unique dual-perspective photocurrent microscope that harnesses the coherent spin dynamics of NV quantum sensors. The high-resolution PCFM here, using magnetic detection\(^{14,23}\), visualizes the photocurrent flow distribution starting at the origin of photoexcitation, whereas electrically detected SPCM characterizes the long-range photocurrent aggregated at the contact. These synergistic viewpoints revealed that thermoelectric anisotropy can stimulate the collected photocurrent in type-II Weyl semimetals when symmetries are broken by sample edges or device-contact placement. Crucially, our work suggests guidelines to distinguish APTE from nonlinear photocurrents in SPCM experiments. Since \( J_{\text{ph}} \) for the nonlinear response should be unidirectional within the beam spot, it can be collected in the device interior even for uniform \( \nabla \psi \) created by extended line contacts.

In devices with spatially varying \( \nabla \psi \), photocurrents due to APTE can be collected over extended bulk areas, in contrast to other mechanisms, including those possessing rotational symmetry\(^{24,29,30}\), whose collection is restricted to interfaces or edges\(^{15,17}\). Moreover, unlike nonlinear photocurrents that exhibit a peaked response at certain photon energies\(^{13,50}\), the APTE is effective for broad wavelength bands through the material’s optical absorption and may appear in a wide range of crystal structures. In Extended Data Fig. 7, we propose prototype device designs based on APTE that enhance the collection efficiency for bulk photocurrents and enable four-quadrant, position-sensitive photodetection in a single chip.

Our visualization of spontaneous vortical currents in thermoelectrically anisotropic materials may also be relevant to experiments on similar materials where temperature gradients could arise, for example, by resistive heating. An intriguing extension is to explore photocurrent in the hydrodynamic regime of WTe₂ (refs. 35, 51). Moreover, our dual-beam quantum magnetometry technique opens the non-contact measurement of thermoelectric effects in anisotropic superconductors, where the interplay between quasiparticle current and supercurrent counterflow may engender interesting spatial distributions\(^{45}\).
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Methods

Sample details
High-quality crystals of WTe₂ are grown in a multistep process. First, a powder mixture of WTe₂ is synthesized by heating a stoichiometric mixture of W and Te (both with 99.999% purity) inside an evacuated silica tube. The mixture is heated to 450 °C for 24 h and 800 °C for another 24 h at the rate of 1 °C min⁻¹, followed by quenching in water. Second, a mixture of powdered WTe₂ (0.2 g) and Te (10.0 g) is heated to 825 °C at 2 °C min⁻¹, held for 2 days, cooled to 325 °C at 4 °C °C⁻¹ and centrifuged to remove the excess Te flux. Third, we anneal the flux-grown samples under vacuum in a two-zone furnace with the hot end at 415 °C and cold end at 200 °C for 2 days to remove any remaining Te impurity and reduce defects.

Bulk TaIrTe₄ crystals are synthesized via the self-flux method. A total of 2.0 g powder with a molar ratio of Ta:Ir:Te = 1:1:12 are loaded in a silica tube and sealed under a high-vacuum condition (<10⁻⁶ Pa). Then, the silica tube is heated to 1,000 °C within 3 days and held at this temperature for 7 days. Finally, the tube is cooled down to 600 °C within 21 days. Needle-shaped TaIrTe₄ crystals are picked out from the ingot.

WTe₂ and TaIrTe₄ flakes are exfoliated onto a polydimethylsiloxane stamp, which is then pressed onto and slowly peeled away from the diamond membrane to transfer the target flakes. The diamond membrane used in this work is cut and polished to 100 μm thickness (Delaware Diamond Knives) from a bulk electronic-grade diamond sample grown by chemical vapour deposition (Element Six). An NV ensemble is created in the diamond membrane by ¹⁵N-ion implantation at 45 keV energy and an areal dose of 10¹² ions cm⁻², followed by annealing at 1,050 °C for 2 h. The XY8-2 sequence acts as the quantum-mechanical analogue to conventional phase-sensitive lock-in detection of photocurrents due to a modulated light source ¹⁰,¹¹,¹⁷,⁵³. Here delay ∆θ is the mean precession angle for delay θ. We empirically determine that a small delay θ = 0.11π rad that is constant over the full imaging region leads to favourable image quality.

NV centre measurements
The diamond membrane with the photocurrent sample facing up (Fig. 1a) is mounted onto a quartz slide. The quartz slide features a lithographically defined coplanar waveguide to deliver the microwave pulses for manipulating the NV centre spin. The NV readout (515 nm) and photocurrent (661 nm) lasers, both linearly polarized, pass through the quartz slide and diamond membrane to impinge on the NV ensemble layer and adjacent photocurrent sample at the top diamond surface. PCFM imaging is performed with 370 μW green laser power and a d.c. photon count rate of 5 × 10⁶ counts s⁻¹ for the NV ensemble. As an example, the PCFM image (Fig. 2a) acquires 35 × 35 pixels with 500 nm resolution and 67 s integration time per pixel for a total frame time of 23 h.

To determine ∆φ at each pixel, we perform an XY8-2 sequence with pulse spacing τ = 6.6 μs and total NV-centre free precession time 16τ = 105.6 μs, delayed by θ = 0.11π rad relative to the pulse photocexcitation ⁶⁻¹⁽¹⁰⁻¹⁴⁻¹⁾. The XY8-2 sequence acts as the quantum-mechanical analogue to conventional phase-sensitive lock-in detection of photocurrents due to a modulated light source ¹⁰,¹¹,¹⁷,⁵³. Here delay θ is the mean precession angle for delay θ. The total photon integration time at each pixel is divided into four different final projection pulses: X₁₁/₂ and Y₁₁/₂. Where, for example, X₁₁/₂ denotes a rotation of the NV superposition state |ψ⟩ around the X axis of the Bloch sphere by +π/2 rad. After each projection pulse, the NV photon count rate (PL) during the green-laser readout pulse is recorded, and we determine the differential projections Xₜ = PL(X₁₁/₂) − PL(X₁1/₂) and Yₜ = PL(Y₁₁/₂) − PL(Y₁₁/₂) to reject common-mode noise. The accumulated phase ∆φ with respect to the initial NV state |ψ⟩₀ = (|0⟩ − |−1⟩)/√2 prepared by an X₁₁/₂ pulse is then computed as ∆φ = arctan(Yₜ/Xₜ).

We set the photocurrent excitation power P such that ∆φ in the PCFM image approximately fills the range ±π, which improves the signal-to-noise ratio but still avoids phase-unwrapping ambiguities. Higher P increases unintentional excitation of the NV centre during the XY8-2 sequence and reduces readout contrast. In addition, we utilize a small delay θ = 0.11π rad between the microwave spin-driving and photocurrent excitation pulses to mitigate the effect of phase broadening over the probed NV-centre spot size. If the photocurrent magnetic field Bₐ is non-uniform over the NV spot size, the coherence of the final NV state |ψ⟩ is damped by a factor exp(−σ₂Δφ(∆φ(θ)/Δφ(0))²/2) where σ₂ is the variance in the phase precession angle over the probed NV spot and Δφ(θ) is the mean precession angle for delay θ (ref. ⁵³). Optimizing the sensitivity, thus, requires balancing the amplitude of phase precession Δφ(θ) (which decreases for larger θ; Fig. 2a) with the contrast of readout (which increases for larger θ). We empirically determine that a small delay θ = 0.11π rad that is constant over the full imaging region leads to favourable image quality.

Anisotropic thermoelectric transport
In this section, we prove that the solution J(r) to anisotropic Seebeck transport equations ¹⁵ (equation (1)) is linear in S_a − S_b. The steady-state condition V·J = 0 imposes the second-order linear partial differential equation for the electrochemical potential φ:

$$\sigma_a \partial_x^2 \phi + \sigma_b \partial_y^2 \phi = -\left(S_a \sigma_a \partial_x^2 T + S_b \sigma_b \partial_y^2 T\right)$$  \hspace{1cm} (3)

with boundary equation

$$n \cdot J = 0$$  \hspace{1cm} (4)

on the sample edges. Since equation (3) is linear, we separate the source term (right-hand side of equation (3)) into two parts and look for solutions of the form φ = φ_a + φ_b, where

$$\sigma_a \partial_x^2 \phi_a + \sigma_b \partial_y^2 \phi_a = -S_a (\sigma_a \partial_x^2 T + \sigma_b \partial_y^2 T)$$  \hspace{1cm} (5)

and

$$\sigma_a \partial_x^2 \phi_b + \sigma_b \partial_y^2 \phi_b = -(S_a - S_b) \sigma_a \partial_x^2 T. \hspace{1cm} (6)$$

We choose the coefficients corresponding to the two potentials φ_a,b to be

$$J_{a,x} = -\sigma_a (\partial_x \phi_a + S_b \partial_y T)$$  \hspace{1cm} (7)

and

$$J_{b,x} = -\sigma_a \partial_x \phi_b. \hspace{1cm} (8)$$

respectively, such that the total current J = J_a + J_b is unchanged from the original problem (equation (1)).

Looking only at equation (5) (isotropic part), we have the simple solution φ_a = −S_a T. This yields the current as

$$J_{a,x} = -\sigma_a (S_a - S_b) \partial_x T$$  \hspace{1cm} (9)

Now, the remaining problem is to solve for φ_b under equation (6) (anisotropic part), subject to the boundary equation on the total current J (equation (4)), which implies that

$$n \cdot J_b = -n \cdot J_a. \hspace{1cm} (10)$$

Since J_a = S_a − S_b (equation (9)), it is clear that both equations (6) and (10) are linear in S_a − S_b. Hence, the solution φ_b and accordingly J_b must also be proportional to S_a − S_b. Thus, the total current J = J_a + J_b = S_a − S_b.

Ab initio calculations of anisotropic in-plane thermopower
Ab initio calculations are performed within the framework of...
density functional theory calculations, including spin–orbit coupling implemented in the Vienna ab initio simulation package (VASP). We use the Perdew–Burke–Ernzerhof exchange–correlation functional and the projector augmented-wave method with an energy cutoff of 312 eV. The ground state is determined on a $12 \times 10 \times 6$ Γ-centred $k$ grid with a Gaussian smearing of 0.05 eV. A subsequent non-self-consistent calculation is employed to determine the band energies on a fine $k$ grid of $144 \times 120 \times 72$, which is used in the transport calculations.

The Seebeck coefficients of WTe$_2$ along the in-plane $a$ and $b$ axes, namely, $S_a$ and $S_b$, respectively, are calculated within the constant relaxation-time approximation for carrier lifetimes, which has been previously found to accurately describe the thermopower of a number of materials.

Then, the temperature-dependent thermopower can be expressed as

$$S_i(T) = -\frac{1}{eT} \int \frac{a_i(\varepsilon)(\varepsilon - \mu)}{\frac{\partial f}{\partial \varepsilon}} \, d\varepsilon,$$

where the transport distribution function is

$$a_i(\varepsilon) = \sum_j \int d\varepsilon v_n^0(k) \delta(\varepsilon - \varepsilon_n(k)).$$

Here $T$ is the temperature, $\varepsilon$ is the carrier energy, $f$ is the Fermi distribution function, $e_n^0(k)$ gives the energy in band $n$ with wavevector $k$, for which $v_n^0(k)$ is the $i$th component of velocity ($i = a$ or $b$). All the energies are measured from the temperature-dependent chemical potential $\mu(T)$, and the $\delta$-function in equation (12) is handled with the tetrahedron method.

Calculations are performed using the experimental lattice parameters: $a = 3.477$ Å, $b = 6.249$ Å and $c = 14.018$ Å (ref. 53). The band structure, Fermi surface and density of states near the Fermi level of WTe$_2$ are shown in Extended Data Fig. 3a–c. The band structure is nearly identical to that obtained in prior first-principles calculations. The calculated $S_a$ and $S_b$ values are plotted as a function of temperature $T$ (Extended Data Fig. 3d). Around room temperature, both $S_a$ and $S_b$ are negative, reflecting larger contributions from electrons in the low-lying conduction bands compared with the corresponding contributions from holes occupying the uppermost valence bands. A temperature-dependent anisotropy is evident; for example, $|S_a|$ is approximately five times larger than $|S_b|$ at 300 K. For $T$ increasing above 300 K, $S_a$ becomes more negative, whereas $S_b$ changes sign and becomes increasingly positive. In Supplementary Section 6, we comprehensively discuss the balance between electron and hole contributions to the thermopower that underlie its temperature-dependent behaviour.

Data availability
The data that support the findings of this study are available from the corresponding author upon request. Source data are provided with this paper.

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Author contributions
Y.-X.W. and B.B.Z. devised the experiments. Y.-X.W. fabricated the photocurrent devices and performed the SPCM and PCFM experiments. Y.R. conceived and demonstrated the theoretical model, with final implementation by Y.-X.W. C.L. and D.B. performed the ab initio calculations. Y.X. and F.T. synthesized the WTe$_2$ samples. R.D. and X.Y. performed the SPCFs and PCFs and the photocurrent devices. Y.-X.W. and B.B.Z analysed the experiments. Y.R. acknowledges support from the NSF award no. ECCS-0335765. CNS is part of Harvard University.

Competing interests
The authors declare no competing interests.

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Extended Data Fig. 1 | Coincident SPCM and PCFM imaging of WTe$_2$ Device B.

(a) Experimental SPCM image of $I_{\text{global}}$ in WTe$_2$ Device B for $P = 100 \, \mu\text{W}$ at 661 nm. (b) Experimental SPCM image of $I_{\text{global}}$ for $P = 300 \, \mu\text{W}$ at 515 nm. The collected photocurrents have identical spatial patterns regardless of optical power or wavelength (see also Fig. 4a of main text). The intensity is slightly higher at 515 nm likely due to better objective transmission and sample absorption at shorter wavelengths. (c) The streamlines for the weighting field $\nabla \psi$ within Device B. We solve $\nabla^2 \psi = 0$ with equipotential surfaces $\psi = 1$ and $\psi = 0$ around two localized positions on the upper (current collecting) and lower (grounded) electrical contacts, respectively. The background colormap denotes the potential $\psi$. (d) Linear dependence of $I_{\text{global}}$ on the optical power $P$ at 661 nm for fixed photoexcitation locations on the $\langle 1\overline{1}0 \rangle$ (red) or $\langle 110 \rangle$ (blue) edges in Device B. The global electrical measurement is consistent with the local magnetic measurement ($\Delta \phi$) presented in Fig. 2b of the main text. (e) PCFM image of $J(r)$ when photoexciting the region of high interior $I_{\text{global}}$ near the upper gold (Au) electrical pad. The four-fold APTE pattern is negligibly changed from the center of the flake. (f) PCFM image for photoexcitation at the corner between the $\langle 110 \rangle$ and $\langle 1\overline{1}0 \rangle$ edges. The locations for the measurements (d-f) within Device B are labeled in the SCPM image shown in a).
Extended Data Fig. 2 | Absence of any detectable photo-Nernst effect in WTe₂ at room temperature. (a) NV center phase image \( \Delta \varphi (r) \) in WTe₂ Device A for an external dc magnetic field, \( B_{\text{ext}} = +28.2 \text{ mT} \). (b) \( \Delta \varphi (r) \) image for the opposite field direction, \( B_{\text{ext}} = -28.2 \text{ mT} \). By flipping the direction of \( B_{\text{ext}} \), we probe the projection of the photocurrent’s magnetic field, \( B_{\text{PC}} \), along opposite NV axes. Afterwards, the NV center’s acquired phase simply changes sign everywhere, indicating that the current flow is independent of the magnetic field direction. (c) Reconstructed \( \mathbf{J}(r) \) for \( B_{\text{ext}} = +28.2 \text{ mT} \). (d) Reconstructed \( \mathbf{J}(r) \) for \( B_{\text{ext}} = -28.2 \text{ mT} \). (e) The difference between the experimental photocurrent patterns at positive and negative external field: \( \mathbf{J}(r, +B_{\text{ext}}) - \mathbf{J}(r, -B_{\text{ext}}) \). The difference highlights the field-antisymmetric component of the photocurrent flow, which isolates the photo-Nernst effect\(^{1,29}\). The overlaid false colormap denotes the magnitude of the difference, which is experimentally consistent with noise. (f,g) Simulated photocurrent pattern \( \mathbf{J}(r) \) when including both the APTE term and a possible Nernst term \( \propto B_{\text{ext}} \hat{z} \times \nabla T \) for f) positive and g) negative \( B_{\text{ext}} \) (see Supplementary Section 7). (h) The difference \( \mathbf{J}(r, +B_{\text{ext}}) - \mathbf{J}(r, -B_{\text{ext}}) \) between the simulated patterns clearly shows a chiral photocurrent vortex due to the photo-Nernst effect, which is absent in the experiment. The colormaps in (f-h) denote the magnitude \( |\mathbf{J}(r)| \).
**Extended Data Fig. 3** | *Ab initio calculations of the band structure and in-plane thermopower of WT$_2$Te$_2*. (a) Electronic band structure calculated within density functional theory, including spin-orbit coupling. (b) Fermi surface of WT$_2$Te$_2$ with an outer electron pocket shown in cyan and an outer hole pocket shown in purple. (c) Electronic density of states for WT$_2$Te$_2$, displaying an electron-hole asymmetry about $E = 0$, defined at the chemical potential $\mu(300$ K). (d) Calculated Seebeck coefficients $S_a$ and $S_b$ along the $a$- and $b$-axes of WT$_2$Te$_2$ versus temperature $T$. Both $S_a$ and $S_b$ are negative at 300 K, with $S_a < S_b$ as observed in experiment. The difference $|S_a - S_b|$ diverges with increasing temperature up to 500 K; the thermopower along the $a$-axis is dominated by electron contributions due to the high velocities of the low-lying conduction bands along this direction, but the smaller velocities along the $b$-axis do not compensate for the increasing hole contribution that is probed at higher temperatures, due to the rapidly rising density of states on the hole side away from the chemical potential.
Extended Data Fig. 4 | APTE photocurrent in TaIrTe₄ Devices A and B. (a) Optical micrograph of TaIrTe₄ Device A, with thickness 440 nm. (b) NV center phase image \( \Delta \phi(r) \) for photoexcitation in the interior of TaIrTe₄ with \( P = 90 \, \mu \text{W} \). The reconstructed \( \mathbf{J}(r) \) from \( \Delta \phi(r) \) is shown in Fig. 3d of the main text. (c) Phase image \( \Delta \phi(r) \) for photoexcitation of the (100) edge in TaIrTe₄ with \( P = 90 \, \mu \text{W} \). The photoexcitation locations of b) and c) are labeled in the optical micrograph shown in a). (d) Reconstructed PCFM image of \( \mathbf{J}(r) \) near the (100) edge, corresponding to \( \Delta \phi(r) \) in c). (e) Dependence of the edge photocurrent \( I_{\text{global}} \) on the optical power \( P \) at 661 nm for an oblique (110) edge in TaIrTe₄. The data are taken on TaIrTe₄ Device B with thickness of 280 nm, whose optical micrograph is shown as the inset. (f) Experimental SPCM image of TaIrTe₄ Device B. Similar to WTe₂, global edge photocurrent is detected along the right (110) edge, while interior photocurrents are detected throughout the device. Far away from the contacts, the edge photocurrent is nearly vanishing along the left (100) edge, as expected from mirror symmetry and a gradient field \( \nabla \psi \) that becomes slowly-varying over the local photocurrent extent. (g,h) PCFM image of \( \mathbf{J}(r) \) for photoexciting: g) the (100) edge, and h) the (110) edge in TaIrTe₄ Device B, both with \( P = 85 \, \mu \text{W} \). The photoexcitation locations for g) and h) are labeled on the SPCM image in f).
Extended Data Fig. 5 | Power dependence of SPCM and PCFM imaging in TaIrTe₄ Device C. (a) Optical micrograph of TaIrTe₄ Device C, with thickness 224 nm. Deliberate point-like side contact is made to the 〈100〉 edge. (b) Calculated weighting field ∇ψ within TaIrTe₄ Device C by assuming point-like equipotential surfaces ψ = 1 and ψ = 0 around the upper and lower locations, respectively, where the gold pads make grazing contact to the sample. c) Full simulation of the SPCM image of the collected photocurrent in Device C using Shockley-Ramo theory. The ∇ψ used is shown in b), and the local $J_{ph}$ is assumed to be due to the APTE with a conductivity anisotropy $\sigma_a/\sigma_b = 4$ for TaIrTe₄ (Fig. 3d,e of the main text). (d,e,f) Experimental SPCM images using 661 nm photoexcitation at powers of 200 μW, 100 μW, and 50 μW, respectively. The spatial pattern is nearly identical across optical powers. g) Linear dependence of the collected $I_{global}$ on the optical power $P$ at three particular locations R1, R2, and R3. The locations of the points are labeled in d). (h,i) PCFM images of the photocurrent flow when exciting the 〈100〉 edge in TaIrTe₄ Device C with $P$ = 100 μW and 50 μW, respectively. The location of the photoexcitation is labeled in e) and f). Although vanishing global photocurrent is collected, as expected from Shockley-Ramo theory for this high-symmetry 〈100〉 edge far from the contacts, the local photocurrent still displays APTE circulation.
Extended Data Fig. 6 | PCFM imaging of edge photocurrent in WTe₂ Device A.
(a) Zoomed-in optical micrograph of WTe₂ Device A. The photoexcitation locations for the PCFM and NV phase images to follow are labeled by their figure panel. (b) PCFM image of $J(r)$ for photoexciting the $\langle 110 \rangle$ edge with $P = 70 \mu W$. (c) PCFM image for photoexciting the $\langle 110 \rangle$ edge with $P = 120 \mu W$. (d) NV center phase map $\Delta \varphi(r)$ for photoexciting the $\langle 100 \rangle$ edge in Device A, corresponding to the reconstructed $J(r)$ shown in Fig. 4d of the main text. (e) Phase map $\Delta \varphi(r)$ corresponding to the $f(r)$ image in b). (f) Phase map $\Delta \varphi(r)$ corresponding to the $f(r)$ image in c).
Extended Data Fig. 7 | Novel photodetector designs based on the APTE. (a) Geometry for enhanced Shockley-Ramo collection of bulk APTE photocurrents. We choose the contact configuration to maximize the dot product between $\nabla \psi$, shown as the streamlines, and $J_{ph}$ due to the APTE in the center of the device. An array of such individual pixels could potentially be used for imaging applications in the mid-infrared or terahertz wavelengths. (b) Simulated $I_{global}$ as a function of the incident beam position for the design in a). The beam size is shown as the green circle and can be realized with an attached microlens. (c) Single-chip, four-quadrant APTE photodetector based on sign-switching, crystal-axes-aligned APTE photocurrents. Photocurrents $I_{13}$, between contacts 1 and 3, or $I_{24}$, between contacts 2 and 4, are alternatively measured with the other contact pair floating. The modeled beam size is shown as the green circle. The material and beam properties are the same between designs shown in b) and c), which allows a comparison of the amplitude of $I_{global}$. (d) The binary pair corresponding to the signs of $I_{13}$ and $I_{24}$ uniquely identifies on which quadrant of the detector the beam is incident. The orientation of the crystal $a/b$-axes for each design is diagrammed.