Photocurrent as a multiphysics diagnostic of quantum materials

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Abstract

The photoexcitation life cycle from incident photon (and creation of photoexcited electron–hole pair) to ultimate extraction of electrical current is a complex multiphysics process spanning across a range of spatiotemporal scales of quantum materials. Photocurrent is sensitive to a myriad of physical processes across these spatiotemporal scales, and over the past decade it has emerged as a versatile probe of electronic states, Bloch band quantum geometry, quantum kinetic processes and device characteristics of quantum materials. This Technical Review outlines the key multiphysics principles of photocurrent diagnostics, for resolving band structure and characterizing topological materials, for disentangling distinct types of carrier scattering that can range from femtosecond to nanosecond timescales and for enabling new types of remote-sensing protocols and photocurrent nanoscopy. These distinctive capabilities underscore photocurrent diagnostics as a novel multiphysics probe for a growing class of quantum materials with properties governed by physics spanning multiple spatiotemporal scales.
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Key points

- The transduction of light into electrical signals (photocurrent) in quantum materials involves physical phenomena across multiple spatiotemporal scales and, therefore, photocurrent stands out as a multiphysics diagnostic tool of quantum materials.
- The long-range collection of locally generated photocurrent, as mediated by the streamlines of diffusion currents, enables a ‘remote’ sensing of local symmetry breaking such as p–n junctions and edges.
- Photocurrent spectroscopy can probe charge, spin and collective excitations with enhanced signal-to-noise characteristics and resolution for atomically thin materials with low optical weight.
- The intimate connection between light–matter interaction and Bloch band quantum geometry renders bulk geometric photocurrents highly sensitive to the crystal symmetry and light polarization.
- Technique innovation including near-field photocurrent probes as well as ultrafast photocurrents grant high spatiotemporal resolution, providing a range of new photocurrent diagnostic tools for quantum materials.

Introduction

The conversion of light (charge-neutral photons) into electrical signals (charged electrons) involves a complex photoexcitation life-cycle process that features a menagerie of physical phenomena across multiple spatiotemporal scales. Photocurrent stands out as a key observable that is active throughout this life cycle, enabling it as an effective sensor of how a quantum material is pushed out of equilibrium by light. Insight into the detailed life cycle of photocurrent has become forthcoming owing to advances in technology, technique and theory over the past decade. For instance, experimental technique innovation (such as time-resolved photocurrent and scanning near-field photocurrent nanoscopy), theoretical advances (such as quantum geometrical origin of bulk photocurrents) and the discovery and engineering of high-quality quantum materials (for example, controlling photocurrents in van der Waals heterostructures) have not only enabled an enhanced understanding of the life cycle but also have used photocurrent as a diagnostic tool.

To track the photoexcitation life cycle, it is useful to consider the various stages of the photoexcitation cascade of an electronic system from initial photoexcitation to carrier relaxation and evolution back to equilibrium. Photocurrent at each stage of this multiscale process can provide snapshots of the electronic quantum state, phase as well as its symmetries. When applied to large-scale material samples or devices, it can also deliver insight into device characteristics. There are four broad stages to the life cycle (Fig. 1). First, there is photoexcitation and absorption, followed by local current generation. After this, the carriers relax and can display a range of complex dynamics. Finally, the photocurrent propagates spatially and can be collected at device contacts. The initial stages of photoexcitation, absorption and current generation occur at the fastest timescales (several to hundreds of femtoseconds), whereas the timescales characterizing carrier relaxation and propagation often depend on material-specific properties and span a wide set of timescales that range from hundreds of femtoseconds to tens of picoseconds and, in some materials, even longer timescales. We note that these latter processes (relaxation and propagation) can in principle occur in parallel, enabling rich and complex photocurrent dynamics.

The properties of photocurrent during each of these stages contain important information about the electronic state and, as a result, can be used as operational principles for novel photocurrent diagnostics of quantum materials. For example, light–matter interaction and Bloch band quantum geometry are closely related, which can be seen in the optical absorption and current generation processes; this renders photocurrent an ideal probe of band geometry as well as the intricate low-energy features of correlated band structure. Additionally, the sensitivity to the degree in which an electronic distribution is pushed out of equilibrium grants ultrafast photocurrents the ability to disentangle quantum kinetic and relaxational processes. Ultrafast autocorrelation techniques and time-domain photocurrent effectively use this distinct sensitivity. Photocurrents can also possess a spatially non-local and long-range character; this permits a ‘remote’ sensing of local photoinduced currents generated far away from current-collecting contacts (a type of photocurrent spatial diagnostics).

In this Technical Review, we highlight the multiscale and multiphysics life cycle of photocurrent in quantum materials, with a particular focus on the key principles of how photocurrent at each stage of the life cycle can be used as a sensitive probe of the out-of-equilibrium properties of quantum materials.

Photocurrent collection

A spatially local photoinduced current (local photocurrent) generated in a tight laser beam spot can be sensed far away at remote current-collecting–voltage-sensing contacts (Fig. 2a). Strikingly, this remote sensing can occur over distances far larger than the electron mean-free path of the device; the primary photoexcited carriers do not have to propagate ballistically to reach the contacts in order for photocurrent to be collected. Its long-range character is a universal characteristic of many opto-electronic devices and is mediated by ambient carriers (those already present in the absence of photoexcitation) in an electronic material. Similar to the way the voltage on a parallel-plate capacitor can be sensitive to the local charge distribution between the plates, the generation of a local photocurrent induces an electromotive force that drives long-range diffusion currents into remote device contacts.

The long-range photocurrent behaviour is often understood using the ‘Shockley–Ramo theorem’ that describes device-specific streamline patterns along which diffusion currents prefer to flow (see Box 1 for description and example streamline patterns). As ambient carriers transmit the signal to remote contacts, the streamlines are agnostic to the precise mechanism of local photocurrent generation. When local photocurrent (a vector quantity) is aligned (anti-aligned) with the streamlines, they produce a maximal and positive (negative) response at the contacts; when local photocurrent is generated in a region with a high (small) density of streamlines, the global current collected at remote contacts is maximized (minimized). This simple framework has been broadly used to account for photocurrent signals in various materials and geometries.

Spatial maps and imaging

Remote sensing of photocurrent enables a versatile diagnostic of spatial features of quantum materials. A typical photocurrent microscopy setup is illustrated in Fig. 2a, in which a tight laser beam spot scans a device sample while the current (or the operationally equivalent
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The photocurrent (photovoltage) is collected at remote contacts—the photocurrent as a function of the position of the laser beam spot is often termed photocurrent spatial maps and enables the imaging of a myriad of spatial features in quantum materials. In these, the spatial resolution is determined by the beam spot size, which is usually about a micron. Because local photocurrent is a vector, photocurrent spatial maps are particularly sensitive to local symmetry breaking. Wang et al. used scanning photocurrent spatial maps to identify local mirror symmetry breaking along edges in a WTe₂ device: along edges where local crystalline mirror symmetry was broken (red boxes in Fig. 2b,c), large photocurrent was measured. In contrast, along edges where crystal mirror symmetry was preserved (orange box, Fig. 2b,d), photocurrent vanished; the photocurrent in the latter maps of Fig. 2d only arose from the contacts. This demonstrates how photocurrent maps can be used to directly probe the topological surface or edge states of a range of topological electronic materials.

Near-field-enabled photocurrent nanoscopy and local probes

When combined with near-field techniques, scanning photocurrent spatial maps can achieve markedly improved resolution. Far-field irradiation incident on an atomic force microscopy probe (Fig. 2e) produces a near-field electromagnetic field at the tip apex that allows a highly localized photoexcitation at length scales of 10–30 nm and thus far below the diffraction limit. Scanning the tip over the sample while probing photocurrent through remote contacts (see inset, Fig. 2e) can reveal spatially localized features of several tens to a hundred nanometres in size. This resolution can be used to image grain boundaries in chemical vapour-deposited graphene, as well as domain walls in twisted bilayer graphene (Fig. 2f). Even as local photocurrent density is generated at highly localized domain walls, the photocurrent collected at device contacts is nevertheless sensitive to the global quantum state, phase, quantum kinetic processes and device characteristics. These snapshots are enabled by novel photocurrent diagnostics (examples given at the top of the figure) that are highly sensitive to electronic degrees of freedom.

Fig. 1 | Multiphysics photocurrent diagnostics probes each stage of the photoexcitation life cycle. Photoexcitation life cycle (labelled 1–4, bottom) consists of broad stages that span multiple spatiotemporal scales. Probing photocurrent during each stage of the life cycle provides distinct snapshots of the electronic state, phase, quantum kinetic processes and device characteristics.
Spatial features such as puddles\(^5\) and, crucially, linking them with device performance as a high-throughput means of identifying other types of localized characteristics and resolution.

Photocurrent as a probe of charge, spin and collective excitations

Photocurrent depends on the transport of electronic degrees of freedom (DOFs), which makes it uniquely sensitive to the charge, spin and collective electronic characteristics of materials. Indeed, even though photocurrent relies on an initial optical excitation, its electrical readout (Fig. 3a) enables the isolation of electronic DOFs; as discussed subsequently, this overcomes experimental and environmental challenges such as low optical weight to achieve enhanced signal-to-noise characteristics and resolution.

In probing electronic DOFs, an important technique is photocurrent spectroscopy (and the related photoconductivity spectroscopy), which is widely applied to measure the absorption spectrum of electronic materials. Both photocurrent and photoconductivity operate in a similar fashion with one key distinction: photoconductivity originates from light-induced changes to the electrical conductivity and requires an external electric bias to generate a measurable photocurrent. In photocurrent spectroscopy, various types of light sources can be used, which include monochromatic sources with continuously tunable wavelengths\(^2\) as well as white sources in which multiple wavelengths are mixed\(^3\). The monochromatic case utilizes high-power wavelength tunable sources, with photocurrent directly measured as a function of wavelength, thereby generating a photocurrent spectrum\(^1\). Sources can take on ranges in the visible\(^2\) and even the terahertz (THz) regimes\(^3\). In using white sources (in the infrared and THz spectroscopy ranges), a Fourier transform methodology can be adopted to obtain the photocurrent spectrum\(^2,3\).

Charge

Although traditionally applied in conventional semiconductors to detect large bandgaps and trapping states in the visible spectrum, photocurrent spectroscopy has recently been successfully used in quantum materials to detect the intricate details of electronic structure and correlated behaviour at low energy scales. Particularly powerful is the Fourier transform infrared photocurrent spectroscopy technique that can realize sensitive infrared and THz spectroscopy even for micron-sized samples of 2D heterostructures\(^2,4,31\). Such photocurrent probes can achieve a...
Box 1
Long-range photocurrent and remote sensing

Spatially localized photocurrent generation and the remote contact sensing in conductors can be captured by a Shockley–Ramo-type approach\textsuperscript{19} that successfully describes the details of various scanning photocurrent spatial maps in quantum materials\textsuperscript{15,16,20–25}. In these maps, a spatially localized photoexcitation (like a tightly focused laser spot) generates a local photocurrent $j_{\text{loc}}(r)$. Even as the localized photoexcitation can be spatially far away from current-collecting contacts, $j_{\text{loc}}(r)$ sets up a local electromotive force that drives diffusion currents $j_{\text{d}}(r) = -\sigma(r) \nabla \phi(r)$ outside the excitation region and into the remote contacts. Here $\sigma(r)$ is the local conductivity tensor and $\phi(r)$ is the electrochemical potential. Current collected at the remote contacts, $I$, follows the simple form\textsuperscript{19}:

$$I = A \int j_{\text{loc}}(r) \cdot \nabla \psi(r) \, dr$$  \hspace{1cm} (1)

where $A$ is a prefactor that depends on device and external circuit configuration, and $\psi(r)$ is a weighting field obtained by solving a Laplace problem $\nabla^2 \psi(r) = 0$ in the device geometry and contact configuration in which photocurrent is measured. Although Eq. (1) describes photocurrent in gapless materials\textsuperscript{19}, long-range Shockley–Ramo type responses persist even in the absence of ambient carriers (like in insulators or vacuum-tube electronics) in which the original Shockley–Ramo theorem\textsuperscript{20,21} applies. The form of the Shockley–Ramo response is ubiquitous and arises under general reciprocity considerations\textsuperscript{8,127}.

From an intuitive perspective, Eq. (1) arises from the incompressibility of the total current density $j_{\text{tot}}(r) = j_{\text{loc}}(r) + j_{\text{d}}(r)$ in the steady state; $j_{\text{loc}}(r)$ generated locally induces $j_{\text{d}}(r)$ far away. This evokes a simple picture of the streamlines $\nabla \psi(r)$: they trace out the lines diffusion currents prefer to follow and do not terminate in the bulk, but flow from one contact into the ground. This enables $I$ to sense the direction of $j_{\text{loc}}(r)$. As an example, consider the simple two-terminal rectangular geometry in which such streamlines flow uniformly from one contact into the ground; this pattern mirrors that of an analogous Laplace problem — that of electric field lines in a parallel-plate capacitor. Local photocurrent generated far away from contacts can be sensed remotely by $I$, with a sign determined by whether local photocurrent $j_{\text{loc}}(r)$ is aligned or anti-aligned with the streamline pattern $\nabla \psi(r)$. For example, local photocurrent generated in the bulk around $n$-doped areas enables the identification of $p$–$n$ interfaces (panel a); similarly, when local photocurrent is generated along edges (such as in topological materials), it also can be detected remotely panel b. Note the strength of $I$ depends on the density of the streamlines allowing strong signals to be collected even from photocurrent generated far from contacts. Streamlines (and hence the responsivity) can be contorted by the device geometry (panel c) and by introducing floating (non-current-collecting) contacts (panel d), as well as by toggling contact configuration that can be used to infer local photocurrent direction.

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**Figure Legend:**

**Device geometry**
- Sample with $p$–$n$ interfaces
- Sample with edge currents

**Photocurrent map**
- Sample with $p$–$n$ interfaces
- Sample with edge currents

**Streamline patterns**
- Sample with $p$–$n$ interfaces
- Sample with edge currents

**A** Sample with $p$–$n$ interfaces
- $I_{\text{loc}}$
- $I$

**B** Sample with edge currents
- $I_{\text{loc}}$
- $I$

**C** Patterned device geometry
- $I_{\text{loc}}$
- $I$

**D** Floating contacts
- $I_{\text{loc}}$
- $I$

**Notes:**
- $j_{\text{loc}}(r)$ represents the local photocurrent generated locally.
- $j_{\text{d}}(r) = -\sigma(r) \nabla \phi(r)$ represents the diffusion current.
- $\sigma(r)$ is the local conductivity tensor.
- $\phi(r)$ is the electrochemical potential.
- $A$ is a prefactor that depends on device and external circuit configuration.
- $\psi(r)$ is a weighting field obtained by solving a Laplace problem $\nabla^2 \psi(r) = 0$.
- $I$ is the current collected at the remote contacts.

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**References:**
- [15,16,20–25]
- [8,127]
Photocurrent measurements of charge, spin and collective excitations in quantum systems. 

**a.** Illustration of a dual-gated ABC trilayer graphene (TLG)–hexagonal boron nitride (hBN) moiré system with a moiré wavelength of 15 nm. 

**b.** Calculated band structure of the moiré system in part a. Dashed curves correspond to the band structure at zero displacement field, whereas solid curves correspond to the band structure at finite displacement field. Arrows (\(I_1\) to \(I_4\)) show the possible interband optical transition at the finite displacement field. 

**c.** Photocurrent infrared spectroscopy can probe the charge excitations in the moiré system. Owing to the low photon energies and high signal-to-noise ratio, photocurrent infrared spectroscopy can resolve tiny energy gaps. The prominent photocurrent peak at \(-72\) meV corresponds to the optical transition \(I_1\) and the peak indicated by green arrows corresponds to \(I_2\) and \(I_4\). 

**d.** Illustration of the non-radiative energy transfer (NRET) between the excited nitrogen vacancies (NV) centre and the nearby graphene and the transient photocurrent. The microwave (MW) signal to drive electron spin resonance (ESR) of the NV centre is applied to two striplines that act as near-field antennas and propagate currents as transient electromagnetic waves (inset). 

**e.** Schematic of the allowed optical transition in nitrogen vacancies and energy transfer processes to graphene. f. Detection of the dynamics of quantum spins in nitrogen vacancies through energy transfer into graphene probed by ultrafast photocurrent mapping. g. Illustration of the scanning near-field photocurrent setup on a boron nitride-encapsulated graphene p–n junction device with split local gates (labelled as LG1 and LG2). The nanotip is scanned across the entire sample and an electrical current (photocurrent) \(I_{\text{PC}}\) is measured simultaneously. h. Local heating at the p–n junction caused by the terahertz nanotip generates a photothermoelectric effect (PTE) current. 

**i.** Photocurrent generated by a PTE owing to the absorption of terahertz collective modes. Intriguingly, near the p–n junction, there are photocurrent oscillations that are perpendicular to the graphene edge. Those oscillations reveal the excitation of propagating surface plasmons. 

Fig. 3 | Much higher signal-to-noise ratio (when compared with purely optical methods) as the electrical current readout does not pick up the environmental optical background. Indeed, a recent Fourier transform infrared photocurrent spectroscopy applied the technique on trilayer graphene-boron nitride moiré superlattices to resolve tiny moiré miniband gaps as well as emergent correlated charge gaps (Fig. 3b,c).
In addition to probing highly excited states, photocurrent can also be sensitive to the details of the electronic Fermi surface, especially in 2D metals and semimetals in which photocurrent generation is often dominated by the photothermoelectric effect (PTE). A strong example is graphene in which the small electronic heat capacity and weak electron–phonon coupling lead to significant heating of the electronic system after optical absorption \(^{36}\). Elevated electronic temperatures at a p–n junction can drive hot electrons and holes in opposite directions, generating a PTE current \(^{22,26,27,37}\). Such photocurrent is highly sensitive to the Fermi surface. For instance, as the PTE is controlled by the Seebeck coefficient that in turn depends on the Fermi surface (both size and polarity), PC can be highly sensitive to the local chemical potential enabling the identification of charge puddles \(^{5}\) as well as time-reversal symmetry breaking through a Nernst effect \(^{16}\). From a broader point of view, PTE can be used as a thermal probe for quantum optical non-local effects was demonstrated \(^{47}\), through the observation of propagating THz plasmons while photocurrents that are sensitive to different aspects of the Bloch band can be used to detect excitation and dynamics in other quantum systems such as spins in quantum defects. As discussed in ref. \(^{17}\), in a setup consisting of nitrogen-vacancy quantum emitters close to graphene, after optical excitation of the NV centres, the relaxation occurs both through radiative channels (luminescence) and non-radiative energy transfer into graphene, which is spin-dependent for NV centres. The latter excites electron–hole pairs in graphene, which can be converted into electrical signals directly or via decay into hot carriers to generate a photothermoelectric current. By measuring the temporal dynamics of the photocurrent in the NV–graphene hybrid system, it was found that the photocurrent dynamics is shorter than the intrinsic NV lifetime but longer than the graphene carrier dynamics. This renders photocurrent a novel electrical readout method of quantum spin transitions. This technique can be used to detect localized photocurrent generation \(^{48}\). In spin–orbit-coupled systems, photocurrent can even be used as a means of controlling spin: circular-polarized light can induce spin-polarized photocurrents \(^{49,50}\).

### Spin

Beyond detecting charge DOFs, PC can additionally be sensitive to a range of other electronic DOFs. For instance, via an energy transfer process between graphene and other quantum systems, the PTE can be used to detect excitation and dynamics in other quantum systems such as spins in quantum defects. As discussed in ref. \(^{17}\). Satellite TO phonons is a well-defined direction in real space), and its generation requires a p–n junction, in bulk photovoltaic effect, photocurrents are generated at macroscopic p–n junctions, in bulk photovoltaic effect, photocurrents are generated uniformly throughout the bulk of a material in the absence of p–n junctions. Such bulk photocurrents possess a direction that depends on a combination of factors that include the polarization, quantum geometry linking quantum geometrical quantities such as the quantum metric and Berry curvature to opto-electronics. Such quantum geometric quantities characterize the texture of Bloch states in momentum space: for instance, the quantum metric describes the distance between two Bloch states close to each other in momentum space. Similarly, the Berry curvature tracks the geometric phase that electrons accrue as they evolve in a small loop in momentum space. Indeed, optical selection rules for photoexcitation often arise hand-in-hand with non-trivial quantum geometry. A prominent example is that of valley-selective circular dichroism in gapped Dirac materials such as gapped bilayer graphene or transition metal dichalcogenides; the opposite signs of Berry curvature in K and K’ valleys enable circularly polarized light to selectively photoexcite electrons in distinct valleys granting access to valley opto-electronics such as a photinduced valley-selective Hall conductivity.

### Quantum geometric photocurrents

At first glance, Bloch band quantum geometry and light–matter interaction may seem unrelated. The former characterizes how Bloch electronic states evolve as a function of momentum \(^{54,55}\), whereas the latter (and its concomitant photocurrents) are often described within an electric–dipole interaction \(^{18}\). Recently, however, theory \(^{57–60}\) has emphasized their intimate relationship: the same dipole matrix elements that describe the transition amplitudes between initial and final states during photoexcitation also form the basis for an interband quantum geometry linking quantum geometrical quantities such as the quantum metric and Berry curvature to opto-electronics. Such quantum geometric quantities characterize the texture of Bloch states in momentum space: for instance, the quantum metric describes the distance between two Bloch states close to each other in momentum space. Similarly, the Berry curvature tracks the geometric phase that electrons accrue as they evolve in a small loop in momentum space. Indeed, optical selection rules for photoexcitation often arise hand-in-hand with non-trivial quantum geometry. A prominent example is that of valley-selective circular dichroism in gapped Dirac materials such as gapped bilayer graphene or transition metal dichalcogenides; the opposite signs of Berry curvature in K and K’ valleys enable circularly polarized light to selectively photoexcite electrons in distinct valleys granting access to valley opto-electronics such as a photinduced valley-selective Hall conductivity.

### Photocurrent diagnostics of quantum geometry

Quantum geometry can manifest in opto-electronic phenomena far beyond that of optical absorption \(^{12,13}\). A striking example is the generation of bulk photocurrents in non-centrosymmetric materials, the so-called bulk photovoltaic effect \(^{56–60,62–64}\). Unlike conventional photovoltaics for which photocurrents are generated at macroscopic p–n junctions, in bulk photovoltaic effect, photocurrents are generated uniformly throughout the bulk of a material in the absence of p–n junctions. Such bulk photocurrents possess a direction that depends on a combination of factors that include the polarization, quantum geometry of the material as well as the presence or absence of symmetries. The last of which is critical as photocurrent is a vector (it adopts a well-defined direction in real space), and its generation requires symmetry-breaking.

There are a number of distinct microscopic mechanisms for bulk photocurrents that are sensitive to different aspects of the Bloch band
Two prominent quantum geometric photocurrents that arise from interband transitions (from an initial to a final state) are injection and shift photocurrents. The injection current arises from the change in carrier group velocity as electrons are photoexcited from an initial to a final state. The shift current, on the contrary, manifests from a real-space shift as electrons are photoexcited. It is often expressed as a shift vector that depends on the polarization of light and captures the real-space displacement when an electron is excited; the shift vector can trace its origins to the geometric Pancharatnam–Berry phase accrued during the transition.

A further recurrent delineation (applied broadly across photocurrent mechanisms) is that of photocurrent responses under specific light polarization conditions, namely, circular or linearly polarized light. This distinction is useful as circular (helicity-dependent) photocurrents and linear photocurrents often generate contrasting behaviour and, under different material symmetry conditions, are either forbidden/allowed (see the table).

A striking example of this distinction are non-magnetic materials wherein only circular injection and the linear shift quantum geometric photocurrents are allowed. The circular injection photocurrent can be understood as follows: the combination of inversion symmetry (P symmetry) breaking and time-reversal symmetry (T symmetry) gives rise to opposite Berry curvature distributions at momenta k and −k, see illustration (red versus blue). These produce optical selection rules that lead to an imbalanced photoexcitation rate for electrons at k and −k under circularly polarized light. The non-uniform k-space photoexcited carrier distribution that ensues possesses carriers with uncompensated group velocity and, therefore, non-zero net current.

The linear shift photocurrent can be understood in an analogous way. P symmetry ensures that the shift vector, characterizing the real space shift between a photoexcited final state and initial state, is an odd function: shift vector at k has an opposite sign from shift vector at −k. When P symmetry is broken, however, this condition is removed allowing optical excitation to induce a shift of the real space position of the charge carrier and a charge current, see wavepacket in illustration. Interestingly, in materials with low enough spatial symmetry, the contribution from different linear polarizations does not compensate with each other enabling non-zero shift photocurrent even under unpolarized light.

In magnetic materials (broken T symmetry), new magnetic photocurrents can emerge. They include the circular shift photocurrent (shifted wavepackets have opposite signs for different helicities: red versus blue wavepackets in illustration) and linear injection photocurrents (excited carriers have uncompensated group velocity). In a special class of magnets, such as PT symmetric antiferromagnets, a composite PT symmetry prohibits the non-magnetic photocurrents discussed earlier, allowing to isolate magnetic photocurrents.

The table below summarizes the different types of quantum geometric photocurrents and their associated symmetry conditions:

| Charge photocurrent | Circular injection | Linear shift | Circular shift | Linear injection |
|---------------------|--------------------|--------------|---------------|-----------------|
| P symmetry          | ×                  |              | ×             |                 |
| T symmetry          | ×                  |              | ×             |                 |
| PT symmetry         | ×                  |              | ×             |                 |

| Material examples   | TaAs, CoSi, RhSi, Td-W(Mo)Te₂ | TaAs, CoSi, RhSi, Td-W(Mo)Te₂ | Even-layered MnBi₄Te₄ | Even-layered MnBi₄Te₄ |
|---------------------|--------------------------------|--------------------------------|-----------------------|-----------------------|
| Microscopy process  | ![Wavepacket Illustration](image1) | ![Wavepacket Illustration](image2) | ![Wavepacket Illustration](image3) | ![Wavepacket Illustration](image4) |

- **Geometric quantity**: Berry curvature (dipole); Shift vector (linear); Pancharatnam–Berry phase
- **Quantum metric (dipole)**

Quantum geometry (see Box 2 for description of bulk photocurrent mechanisms). For instance, for bulk photocurrents arising from interband transitions, circular (linear) injection currents formed from symmetries in the group velocity of the photoexcited carriers are sensitive to the interband Berry curvature (quantum metric)\(^{59,60}\); similarly, shift photocurrents are formed from real space displacements of electrons as they are photoexcited and depend on the shift vector\(^{56,68}\). In non-centrosymmetric metals, bulk photocurrents can even flow when irradiated with light frequencies below the interband optical transition gap. Such metallic photocurrents depend on other quantum geometric quantities such as the intraband Berry curvature dipole\(^{58,59}\) — a full account of these is the subject of current intense investigation\(^{69-74}\). Even in centrosymmetric materials, bulk photocurrents of a quantum geometric nature can be induced by non-vertical transitions mediated by the finite momentum of polaritons\(^{75,76}\).

As a result of the intimate connection between bulk photocurrents and Bloch band quantum geometry, photocurrents have recently emerged as a powerful tool to study topological materials and emergent quantum phases in correlated electronic systems from the perspective of symmetry and quantum geometry. Low photon energies are often key to observing sizable quantum geometric photocurrents arising in topological materials or from correlated electronic systems.
For example, large injection and shift currents were theoretically predicted for infrared and THz photons and experimentally reported by using mid-infrared photons in topological Weyl semimetals TaAs and TaIrTe₄ (refs. 77–79) (Fig. 4a–c) and also a monolayer topological insulator WTe₂ (ref. 80). A particularly interesting prediction is the quantized circular injection rate in Weyl semimetals with spatially chiral lattices in which inversion and mirror symmetries are all broken⁵⁷,⁸¹. Material candidates include CoSi and RhSi in which quantization can be used as a means to empirically count the number of Weyl nodes filled. However, a precise measurement of the current injection rate is challenging owing to the short momentum lifetime of photocarriers. In addition, theory has found that the quantization of CPGE injection in chiral topological semimetals is generally removed by including perturbations from interactions⁸². Nonetheless, several experimental groups have led significant efforts in applying THz time-domain spectroscopy techniques with infrared pump to detect the rare near-quantization phenomenon in non-linear responses⁸³,⁸⁴. Such techniques are described in the following section on ultrafast photocurrent techniques.

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**Fig. 4 | Quantum geometric photocurrents observed in various materials with distinct dimensionalities and symmetry.**

**a.** The polar lattice structure (left) and real picture (right) of the topological Weyl semimetal TaAs crystals. **b.** Illustration of the k-space Berry curvature monopoles in a Weyl semimetal. **c.** The measured circular injection photocurrent that arises from the chirality and Berry curvature of Weyl nodes in TaAs. The circular photocurrent alternates between right-handed circularly polarized (RCP) and left-handed circularly polarized (LCP) light. **d.** Illustration of the crystal lattice of a WS₂ nanotube whose symmetry is lowered owing to reduced dimensionality. **e.** Photocurrent measured as a function of the laser position along the tube. The photocurrent is maximum when the laser is in the bulk of the tube (away from the contacts), indicating its bulk photovoltaic nature. **f.** The photon polarization dependence of the short circuit photocurrent (I_SC) with the laser focused in the middle of the sample. The arrows in the polar plot indicate the direction along the tube for each device. **g.** Illustration of the lattice structure of an engineered two-dimensional heterostructure, twisted double bilayer graphene. **h.** The experimental observation of highly tunable (by light polarization and external electric field) mid-infrared bulk photovoltage (V_ph) in a twisted double bilayer graphene moiré device, arising from quantum geometry in the moiré bands. The light polarization is experimentally controlled by a quarter wave plate (QWP) and the external electric field is controlled by different gate voltages (V_g). Part a adapted with permission from ref. 11, under a Creative Commons licence CC BY 4.0. Parts b and c adapted with permission from ref. 77. Springer Nature Ltd. Parts d–f adapted with permission from ref. 80. Springer Nature Ltd. Parts g and h adapted with permission from ref. 81. Springer Nature Ltd.
Controlling quantum geometrical photocurrents in low dimensions

Given the sensitive dependence of quantum geometrical photocurrents on crystalline symmetries, low-dimensional materials have become a choice platform to control quantum geometrical photocurrents. By exploiting reduced crystalline symmetries in low-dimensional materials (such as nanotubes), enhanced bulk quantum geometrical photocurrents can be realized (Fig. 4d–f). The ability to stack 2D materials together provides a further engineering opportunity — van der Waals interface symmetry engineering. A prime example is that of WSe₂-black phosphorous heterointerfaces. WSe₂ (black phosphorous) possesses threefold (twofold) rotational symmetry. The interface between the two loses all rotational symmetries allowing a large quantum geometrical current to manifest uniformly in the bulk of the sample even when the individual constituents (WSe₂ and black phosphorous) did not exhibit bulk photocurrents. This is a striking manifestation of symmetry engineering and van der Waals controlled photocurrent.

Such engineering capability can be pushed to the extreme in moiré materials where the reconstructed moiré minibands can host pronounced quantum geometrical quantities that depend not only on the twist angle between individual layers but also on device conditions such as applied gate voltage. As such, moiré minibands can produce a complex structure of photocurrent as a function of polarization and gate voltage (Fig. 4g,h). This sensitive dependence on the details of the moiré minibands can be exploited as a means of polarimetry to identify the wavelength and power of the incoming light all in a single moiré device. Looking forward, photocurrent sensitivity to symmetry and wavefunctions may render it capable of diagnosing the menagerie of correlated and spontaneously broken symmetry phases in moiré materials.

Magnetic photocurrents

Beyond spatial symmetries, photocurrent diagnostics can be applied to detect quantum properties under broken time-reversal symmetry. Indeed, broken time-reversal symmetry (in addition to broken inversion) leads to new classes of magnetic photocurrents (see the table, Box 2) that are directly proportional to the magnetic order: when the magnetic order flips, the magnetic photocurrent will also reverse its direction. Two prominent magnetic photocurrents are the circular shift current (shift current induced by circularly polarized light) and linear injection current (injection current induced by linearly polarized light). One special class of magnetic materials that have an urgent need for photocurrent characterization are PT symmetric antiferromagnets. PT symmetry guarantees a vanishing net magnetization making the detection of its antiferromagnetic order a challenging, yet highly desirable, task for spintronic applications. Magnetic photocurrents can provide a non-invasive measure of the antiferromagnetic order and, when combined with scanning techniques, may even enable spatial resolution of magnetic domains.

Although there are a steadily increasing range of PT symmetric antiferromagnetic materials, 2D van der Waals magnets are a particularly simple and illustrative example. For instance, by stacking even layers of the 2D ferromagnet CrI₃, together, spins between adjacent layers become aligned antiferromagnetically realizing a layered PT symmetric antiferromagnetic ground state. In a similar fashion, even layered MnBi₁Te₆ also realizes a layered PT symmetric antiferromagnet. Although both have been predicted to support magnetic photocurrents, MnBi₁Te₆ stands out owing to its non-trivial topology that supports axion electrodynamics.

Ultrafast photocurrents

The quantum kinetics of out-of-equilibrium electronic states can range across a wide multitude of timescales from the femtosecond to picosecond and even nanosecond range. The energy relaxation of a photoexcited weakly interacting electronic system typically proceeds in three broad stages. The first and fastest step is the thermalization of the electron gas by carrier–carrier scattering. In this step, the electrons distribute their energy among each other and can establish an out-of-equilibrium hot-electron distribution that retains the energy of photoexcited carriers. The second stage involves the cooling of the hot electrons, that is, the transfer of the energy out of the electron gas to the lattice. Finally, once carriers are cooled and reach the band edge, the remaining energy is lost through recombination of electron–hole pairs.

Ultrafast photocurrent techniques can be used to probe the quantum kinetics in each of these timescales: each timeframe reveals different particle interactions and scattering processes depending on the exact mechanism and material properties of the system. The techniques combine traditional optical pump probe with various electrical readout schemes that work together to overcome fundamental electronic bandwidth limitations. In contrast to all-optical techniques, which can include contributions from phonons or the substrate, ultrafast photocurrent measurements selectively isolate the electronic system and its associated electronic processes. This allows different (electronic) carrier dynamics and relaxation pathways to be studied depending on the microscopic mechanisms underlying the specific photocurrent generation. For instance, the time evolution of electronic temperature can be probed by photothermoelectric currents, as described subsequently. Similar to photocurrent spectroscopy described earlier, one key advantage of ultrafast photocurrents compared with all-optical pump-probe measurement is its enhanced signal-to-noise quality when applied to small nanoscale materials whose absorption coefficients are weak.

Ultrafast photocurrent autocorrelation

In photocurrent autocorrelation measurements, the steady-state photocurrent is measured under excitation by two time-separated optical pulses. When photocurrent is a non-linear function of intensity, the measurement depends on the pulse separation and allows the photocurrent response time to be inferred. Its operating principle can be understood by considering the total time-integrated photocurrent response produced by two pulsed excitations of equal power $P$ separated by a time $\Delta t$ (Fig. 5a). For a sub-linear photocurrent response, two pulses coinciding in time $\Delta t = 0$ produce a photocurrent that is less than two pulses that are well separated $\Delta t \to \infty$. Hence, the photocurrent response exhibits a dip around $\Delta t = 0$ (Fig. 5b) and saturates as $\Delta t \to \infty$. The saturation point describes the regime where response time is larger than the pulse separation, whereas a small $\Delta t$ describes the intermediate regime where the photocurrent does not have time to completely relax before the second pulse. Therefore, the photocurrent response time can essentially be extracted from the dip in the photocurrent data $PC(\Delta t)$ (Fig. 5b). This technique has proven powerful in probing the electron dynamics in a range of solid-state systems.

The earliest works and application of the technique involved characterization of semiconductor photoconductive switches, in which the entire photocurrent life cycle occurs on picosecond timescales owing to fast carrier cooling and short recombination lifetimes caused by carrier trapping via defects. In low-dimensional systems, however, energy relaxation pathways are strongly attenuated owing to a change in the phase space of quantum kinetic processes. In graphene, for
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Fig. 5 | Ultrafast photocurrents. a, Schematic setup for measuring the autocorrelation of graphene photothermoelectric effect (PTE) current in a dual-gated (bottom gate, BG; top gate, TG) device. b, Graph plots the generated photovoltage as a function of the time delay Δt between two pulses. c, Electron temperature measurements as a function of the time delay tracks the cooling dynamics of the hot-electron gas. The inset illustrates the mechanism for carrier cooling — phonon polariton emission. d, Schematic of the free-space terahertz (THz) emission as a measure of photocurrent in the time domain. e, Generated THz pulses measured from left and right circularly polarized pump light at 45° angle of incidence in a topological chiral semimetal RbSi83. Their difference is the photon helicity-dependent circular photocurrent signal. f, The photocurrent amplitude as a function of the excitation photon energy. g, Schematic of THz time-domain photocurrent spectroscopy. h, Measurement of the helicity dependent photoinduced anomalous Hall current transient as a function of real time measured in graphene. i, Light-induced gap openings in graphene under intense mid-infrared illumination. 

example, slow electron–phonon interactions32,103,104 extend the lifetime of the hot-carrier distribution into the picosecond range within the resolution of ultrafast techniques. Because the photocurrent response in graphene is dominated by its hot-electron distribution (established quickly by fast carrier–carrier thermalization105–108) and the PTE, photocurrent autocorrelation measurements can directly track the cooling dynamics of the hot-electron gas (Fig. 5b,c). In this regard, photocurrent autocorrelation has been used to probe not only the electron–phonon interactions involved in carrier cooling32,109 but even the initial electron–electron scattering-induced thermalization of the hot-electron distribution on femtosecond timescales4. These techniques have even revealed unconventional scattering such as supercollision cooling2 and near-field radiative cooling via phonon polariton modes in remote dielectric layers109 (Fig. 5c).

Although lattice cooling occurs much faster in semiconductors, the presence of an energy gap inhibits rapid recombination, in the absence of defects with large capture cross-sections, so that carriers can reside for some time at the conduction and valence band edge before recombining. Photocurrent response in these systems is instead governed by the diffusion of photoexcited carriers in the presence of electric fields and probes interactions that influence this process. For example, the response time is sensitive to the formation of excitons111.
and their many-body physics, which can be probed using photocurrent autocorrelation\textsuperscript{123–125}.

**Table 1 | Powerful photocurrent diagnostics of spatiotemporal characters enabled by various imaging and spectroscopy techniques**

| Technique | Excitation-probe | Spatial resolution | Time resolution | Enabling technology |
|-----------|------------------|-------------------|----------------|------------------|
| Scanning photocurrent (far-field) | • CW • UV to terahertz | • Diffraction-limited • 1–100 µm | Steady state | Long-range nature of photocurrent enables a remote sensing of spatially localized photoexcitation |
| Scanning photocurrent (near-field) | • CW • UV to terahertz • Focused onto the tip | • Limited by tip size • 10–30 nm | Steady state | Enabled by a nanoscale scanning probe (like an AFM tip) |
| Photocurrent FTIR | • Broadband • UV to terahertz | • Diffraction-limited • ~10–100s of µm | Steady state | Based on FTIR spectroscopy technique; high signal-to-noise ratio than conventional FTIR absorption spectroscopy |
| Photocurrent autocorrelation | • Femtosecond laser • UV to infrared | • Diffraction-limited • ~1–10 µm | 10s of fs | Relies on non-linear photocurrent power dependence |
| NV photocurrent magnetometry | • CW • UV to infrared (excitation) • Visible (probe) | • Diffraction-limited • ~1–10 µm (excitation) • ~1 µm (probe) | Sub-microsecond | Contact-free NV imaging can directly resolve the local photocurrent flow pattern |
| Ultrafast on-chip photocurrent | • Femtosecond • UV to infrared (excitation) • Visible (probe) | • Diffraction-limited • ~1–10 µm (excitation) • ~1 µm (probe) | 10s of fs | Ultrafast temporal resolution that is not limited to non-linear power dependence in photocurrent |

UV, ultraviolet; FTIR, Fourier transform infrared; RF, radio frequency; NV, nitrogen vacancy; CW, continuous wave; AFM, atomic force microscope.

**THz time-domain photocurrent spectroscopy**

THz time-domain photocurrent spectroscopy offers further functionality over the photocurrent autocorrelation technique, in that the photocurrent pulse itself is sampled over ultrafast timescales using electro-optic-based detectors such as ZnTe\textsuperscript{126}. Electro-optic detectors offer the advantage of directly detecting the electric field instead of the intensity, for example, when absorptive photodetectors are used. This technique is routinely used in various settings, such as probing the kinematics in bulk quantum materials, in which an ultrafast pump laser generates transient photocurrents in the crystal, which in turn emits THz radiation that is subsequently detected (Fig. 5d,e). Ultrafast optical pumps are often operated in the visible wavelength range, enabling the study of a vast variety of ultrafast scattering processes and phenomena\textsuperscript{127–129}.

More recently, by tuning the pump photon energy into the infra-red range and studying photocurrent as a function of photon energy, near-quantized plateaus of circular injection current (Fig. 5f) have been directly observed\textsuperscript{83} — a ‘spectroscopic’ signature of the Chern flux of
chiral Weyl semimetals. From a broader perspective, such variable pump-spectroscopy techniques naturally lend themselves in interrogating processes that depend on extremely far-out-of-equilibrium photoexcited distributions (as opposed to hot but thermalized distributions) such as ultrafast spin currents in magnetic heterostructures.

On-chip THz detection
For nanomaterials with weak emission coefficients, THz time-domain spectroscopy can be carried out using an on-chip photoconductive switch. Figure 5g displays a typical schematic of the measurement setup. The photoactive device is contacted between two transmission lines that act as waveguides for the transient photocurrent pulse produced by excitation. At some distance down the transmission lines, a probe pulse is used to open a photoconductive switch that samples the photocurrent on a timescale dictated by the response time of the switch. The photocurrent is measured as a function of the delay time between the pump and probe and directly tracks the magnitude of the photocurrent transient in time. In this manner, the technique allows to study the quantum transport of photoexcited carriers and their out-of-equilibrium phases.

Another striking attribute of ultrafast photocurrent techniques is that they enable the excitation of electronic systems with intensities far higher than sustainable in conventional devices under continuous wave illumination. As such, they provide access to extreme out-of-equilibrium physics that rely on strong instantaneous electric fields. A particularly striking example of this is Floquet band engineering in graphene to produce unconventional Floquet-driven topological phases. By driving graphene with circularly polarized light, theoretical proposals had long predicted an anomalous Hall phase; the requirements on electric field to achieve sizable anomalous Hall effect were challenging: laser intensity requirements exceed those expected to destroy a typical graphene sample under continuous wave irradiation. Instead, McCleer et al. circumvented this by using an ultrafast time-domain photocurrent technique. They achieved large electric fields required for a sizeable anomalous Hall effect in graphene, but only for a short period of time, avoiding damage to the graphene device; here the Auston switch enables the direct measurement of the (Floquet) Hall conductivity (Fig. 5g–i).

Outlook
Photocurrent is a highly versatile and multiphysics diagnostic of a range of electronic processes and properties. Yet, the same sensitivity to processes across multiple scales and stages in the photoexcitation life cycle can sometimes complicate its interpretation. For instance, one challenge is distinguishing between the various mechanisms for photocurrent, for example, differentiating intrinsic photocurrent mechanisms such as quantum geometric photocurrents and “extrinsic” photocurrents that arise at a p–n junction. In this regard, systematically studying the dependence on photon polarization, current direction, excitation wavelength, beam spot location, doping dependence and temporal dynamics can often prove critical in delineating between mechanisms. This is because photocurrent often exhibits distinctive “tell-tale” characteristics: the multiple sign changes of photocurrent as a function of doping density at a graphene p–n junction singled out its photothermoelectric origin; systematic photocurrent spatial maps were used to reveal the bulk origin of photocurrent in WSe$_2$–black phosphorous heterointerfaces. Yet, there is no one-size-fits-all methodology for delineating between mechanisms. Instead, insight into the detailed physics revealed by photocurrent, particularly in completely new material platforms in which little is known, is most often attained when diagnostics are performed hand-in-hand with theory. In the future, combining multiple techniques such as ultrafast optics, polarization control and near-field spatial resolution will lead to next-generation photocurrent setups; used together, these techniques can provide powerful probes not only of photocurrent mechanisms but also means to control out-of-equilibrium physics in a gamut of complex quantum materials, such as moiré materials, all-in-situ.

The multiphysics quantum materials photocurrent diagnostics can be flipped on its head: photocurrent in quantum materials can also be used as highly sensitive polarimeters, photodetectors and sensors. Quantum geometric photocurrents, for instance, have gained particular attention owing to their p–n-junction-less response (an entire sample bulk away from a defined p–n junction can be photocurrent-active), its strong sensitivity to light polarization as well as its sensitivity in the far-infrared and THz regimes. These characteristics have formed the basis for new opto-electronics design concepts. For instance, by measuring circularly polarized light-induced valley Hall photocurrent in gapped bilayer graphene, a photoconductive sensor with suppressed dark current can be achieved. Similarly, pronounced quantum geometric quantities in quantum materials such as moiré heterostructures and Dirac–Weyl semimetals can be used for improved sensitivity in the THz regime where traditional technology has often struggled.

Looking forward, beyond the multiphysics diagnostics areas we described earlier (see Table 1 for a selective summary of key photocurrent diagnostics), photocurrent can also play important roles in research frontiers such as probing-correlated electron systems, optical–dynamical control and their intersection. For example, the PTE current can be used as a probe of insulating gaps in novel correlated systems such as moiré materials, particularly in inhomogeneous samples. This is because the Seebeck coefficient is highly sensitive to resistive states; this contrasts with conventional transport probes that rely on charge current that can be easily shunted along highly conductive regions. Another emerging diagnostic is how geometric photocurrents can be used to probe symmetry breaking and phase transitions owing to its high sensitivity to the symmetry of electronic systems. In particular, the energies of mid-infrared and far-infrared photons often directly match the energy scale of correlated quantum phases, such as charge density waves and Mott gaps, which can lead to significantly increased sensitivity. For instance, recent work used photocurrent as a probe of the unique gyrotropic electronic order in a correlated semimetal TiSe$_2$ (ref. 123). From a dynamical control perspective, ultrafast photocurrents are essentially a light-induced transient electric field; such transient electric fields can be used to control the symmetry of electronic states on the ultrafast timescale. The next-generation photocurrent techniques with nanoscale resolution, femtosecond time resolution and sensitive polarization control will become a multi-scale tool that can interrogate a wide range of novel topological, geometrical and correlated physics.

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