Generation, transport and detection of valley-locked spin photocurrent in WSe$_2$–graphene–Bi$_2$Se$_3$ heterostructures

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Quantum optoelectronic devices capable of isolating a target degree of freedom (DoF) from other DoFs have allowed for new applications in modern information technology. Many works on solid-state spintronics have focused on methods to disentangle the spin DoF from the charge DoF, yet many related issues remain unresolved. Although the recent advent of atomically thin transition metal dichalcogenides (TMDs) has enabled the use of valley pseudospin as an alternative to the spin DoF, it is nontrivial to separate the spin DoF from the valley DoF since the time-reversal valley DoF is intrinsically locked with the spin DoF. Here, we demonstrate lateral TMD–graphene–topological insulator hetero-devices with the possibility of such a DoF-selective measurement. We generate the valley-locked spin DoF via a circular photogalvanic effect in an electric-double-layer WSe$_2$–monolayer graphene–Bi$_2$Se$_3$ heterostructure, and the spin DoF is measured separately in the topological insulator via non-local electrical detection using the characteristic spin-momentum locking. Operating at room temperature, our integrated devices exhibit a non-local spin polarization degree of higher than 0.5, providing the potential for coupled opto-spin–valleytronics that independently exploit the valley and spin DoFs.

Over the past decade, various electrical$^5$, magnetic$^6,7$ and optoelectronic methods$^8$ to decouple the spin DoF from charge DoF have been demonstrated. Manipulating the spin DoF, however, is associated with long-standing issues. For instance, in spin transistors one needs to overcome the resistance mismatch for efficient spin injection/detection$^9$. More importantly, most of spintronic devices operate at cryogenic temperatures to distinguish the spin DoF from the charge DoF$^{10}$, which restricts practical device applications. Owing to their inversion asymmetry and time-reversal symmetry, atomically thin two-dimensional (2D) TMDs exhibit two degenerate valleys near the hexagonal corners of the Brillouin zone. Helicity-dependent optical excitation is currently a routine method to access the valley DoF in these materials$^4$. Since these binary valley DoFs are intrinsically coupled with the spin of the charged carriers, 2D TMDs in principle possess two sets of DoFs—namely the valley and spin DoFs. Although external stimuli, such as high magnetic field$^{11}$, presence of ferromagnetic materials$^{12}$, or optical pseudomagnetic field$^{13}$ can lift the spin–valley locking and thus selectively detect one of the two DoFs, the development of true valleytronic or spintronic devices requires pure electrical detection together with room-temperature operation.

To meet the above requirements, at least the following three processes must be implemented: optoelectronic generation of the valley–spin-coupled DoF, the associated electrical transport, and non-local electrical readout. For the first process, the material must be electrically tunable for generating the coupled valley–spin DoF with a light-helicity-dependent population imbalance between the two valleys. Second, the transport must exhibit weak spin–orbit coupling to maximize the spin diffusion length. Finally, the detection should be implemented to match the spin orientation of electrons across all three materials.

We propose and demonstrate a lateral heterostructure device that consists of three layered heterogeneous materials: WSe$_2$–TMD, monolayer graphene, and Bi$_2$Se$_3$ topological insulator (TI). Our device operates at room temperature and does not require an external magnetic means to control the valley or spin DoFs. Figure 1a presents a schematic of our proposed device, with Fig. 1b and c showing the corresponding side-view and the optical microscopy images (Methods and Supplementary Information I). The device consists of a thick WSe$_2$–TMD flake for optical generation and gate-dependent electrical manipulation of valley-locked spin-polarized electrons, monolayer graphene for the transport channel, and Bi$_2$Se$_3$ TI for non-local detection of the spin-polarized electrons.

We induce an inversion-symmetry-broken Rashba 2D electron gas (2DEG) at the surface of WSe$_2$ by ionic liquid electric-double-layer gating$^{14}$. Valley-locked spin-polarized electrons are excited through the circular photogalvanic effect (CPGE), where the transferred photon angular momentum generates in-plane valley-locked spin DoF electrons. The locally generated carriers diffuse across a submicrometre-long graphene channel. We use monolayer graphene due to its exceptional spin diffusion length ($\sim 2\mu$m)$^{15}$ as well

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as the pseudo-spin nature, with no spin or valley preference for the electrical transport. For detection, we use a non-local, all-electrical detection technique to exploit the unique property of TIs, namely the spin–momentum locking. TIs possess only the in-plane spin texture, and thus the measured polarity of the TI current naturally reflects the valley-locked spin DoF electrons.

Figure 1d–g shows the results of electrical transport measurements. The WSe₂ gate voltage ($V_g$)-dependent drain current ($I_{d,WSe₂}$) shows $n$-type transfer characteristics (red colour in Fig. 1d) with a threshold voltage of 0.75 V. Due to the large WSe₂ resistance, the transfer curve from WSe₂ to Bi₂Se₃ (green colour in Fig. 1d) resembles that of the WSe₂ (red colour in Fig. 1d). Figure 1e–g shows the output characteristics of the WSe₂ transistor, Bi₂Se₃ TI transistor, and WSe₂–graphene–Bi₂Se₃ hetero-device, respectively. The output curve of the WSe₂ transistor measured at $V_g=0$ V exhibits a Schottky behaviour with a resistivity of 50 Ω m (Fig. 1e). The measured resistivity ($198 \times 10^{-4}$ Ω m) for the Bi₂Se₃ TI transistor at a drain voltage ($V_{d,BiSe₃}$) of 0.5 V is low compared to that of WSe₂ due to the metallic character of the Bi₂Se₃ surface states (Fig. 1f). The $I$–$V$ characteristics of the WSe₂–graphene–Bi₂Se₃ device are shown in Fig. 1g.

Figure 2a,b shows the light-helicity-dependent local photocurrent responses for WSe₂ TMD and Bi₂Se₃ TI, respectively. We experimentally exclude the photo-thermoelectric contribution by injecting light exactly at the sign reversal location of each device channel. To transfer the photon angular momentum onto the surface of each device, we tilted the plane-of-incidence (xz plane) to form an oblique incidence angle $\theta$ of 30° with respect to the xy plane. The measured photocurrents exhibit a strong dependence on the polarization of the incident light, and both traces oscillate with the rotation angle $\varphi$ of the quarter-wave plate (QWP). The associated photocurrent $I_p(\varphi)$ can be expressed using the well-known four-component model:

$$I_p(\varphi) = C \sin 2\varphi + L_1 \sin 4\varphi + L_2 \cos 4\varphi + D$$  

Equation (1) includes four photoinduced components: a π-periodic current oscillation term $C \sin 2\varphi$ that corresponds to the CPGE current, a π/2-period oscillation term $L_1 \sin 4\varphi$ that represents a linear photogalvanic effect, and a term $L_2 \cos 4\varphi$ that originates from a linear photon drag effect. Helicity-independent photocurrent is
are applied to each pair of electrodes, 1–2 and 3–4 as shown in Fig. 1a. Red solid lines are the fits to the measured data using equation (1). c. Extracted local CPGE coefficient C is plotted as a function of \( V_g \) with \( V_{C,GSE} = 0 \) V for the WSe\(_2\) TMD. Inset: local CPGE current \( C \) as a function of the WSe\(_2\) gate voltage \( V_g \) for the local and non-local CPGE results. A monotonic increase is observed, similar to the case of local WSe\(_2\) CPGE measurements. To investigate the electronic tunability, we consider the \( V_g \)-dependent polarizability of the valley and spin DoFs. Compared to the relatively straightforward contribution described by the CPGE C coefficient (Fig. 3c), the optically generated polarization degree is rather complicated. Following S. Ganichev et al., we define \( P = (I(\sigma_\uparrow) - I(\sigma_\downarrow)) / (I(\sigma_\uparrow) + I(\sigma_\downarrow)) \), where \( I(\sigma_\uparrow) \) and \( I(\sigma_\downarrow) \) denote photocurrents under left- and right-hand circularly polarized optical excitation, respectively. Combining this with equation (1), we can express \( P \) without the helicity-insensitive contribution as (Supplementary Information II)

\[
P = \frac{|I_l - I_r|}{|I_l| + |I_r|}
\]

Figure 3d,e show the \( V_g \)-dependent \( P \) for local and non-local CPGE measurements, respectively. We observe that \( P \) for the local CPGE result exhibits a linear dependence on \( V_g \), whereas \( P \) for the non-local CPGE increases quadratically. The spin diffusion length of graphene would vary with \( V_g \) because the ionic liquid was deposited on the entire hetero-device. However, previous studies have revealed that the graphene spin diffusion length shows negligible variation with \( V_g \) at room temperature. The spin injection efficiency is very small, of the order of 0.21 % (Supplementary Information II), because of the large momentum mismatch between the three lateral stacks, which poses an upper bound for the \( V_g \)-dependent non-local \( P \).

It is interesting to compare the local \( V_g \)-dependent \( C \) (Fig. 2a) and \( P \) (Fig. 3d) in the WSe\(_2\) transistor. Ab initio calculations have

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**Fig. 2 | Light-helicity-dependent local photocurrent response of WSe\(_2\) and Bi\(_2\)Se\(_3\), a,b, Top:** schematic diagrams for the local helicity-dependent photocurrent in WSe\(_2\) (a) and Bi\(_2\)Se\(_3\) (b). A continuous-wave (cw) diode laser (average power of 10 \( \mu \)W) was used with a photon energy of 1.17 eV, which ensures intra- or intersubband optical transitions. \( \theta \) (30 °) is the incidence angle, and \( \phi \) is the rotation angle of the quarter-wave plate. Bottom: data (filled squares) are the measured \( I_P(\phi) \) with \( V_d = 0 \) V and \( V_g = 1.5 \) V for WSe\(_2\) (a) and Bi\(_2\)Se\(_3\) (b). \( V_{c,WSe2} \) and \( V_{c,Bi2Se3} \) are applied to each pair of electrodes, 1–2 and 3–4 as shown in Fig. 1a. Red solid lines are the fits to the measured data using equation (1).

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**Fig. 3 | Valley-dependent CPGE in WSe\(_2\) and Bi\(_2\)Se\(_3\). a** Top: theoretical calculations for the valley-dependent CPGE in a stack of two layers of WSe\(_2\). Bottom: theoretical calculations for the valley-dependent CPGE in a stack of two layers of Bi\(_2\)Se\(_3\). A continuous-wave (cw) diode laser (average power of 10 \( \mu \)W) was used with a photon energy of 1.17 eV, which ensures intra- or intersubband optical transitions. \( \theta \) (30 °) is the incidence angle, and \( \phi \) is the rotation angle of the quarter-wave plate. Bottom: data (filled squares) are the measured \( I_P(\phi) \) with \( V_d = 0 \) V and \( V_g = 1.5 \) V for WSe\(_2\) (a) and Bi\(_2\)Se\(_3\) (b). \( V_{c,WSe2} \) and \( V_{c,Bi2Se3} \) are applied to each pair of electrodes, 1–2 and 3–4 as shown in Fig. 1a. Red solid lines are the fits to the measured data using equation (1). c. Extracted local CPGE coefficient C is plotted as a function of \( V_g \) with \( V_{C,GSE} = 0 \) V for the WSe\(_2\) TMD. Inset: local CPGE current \( C \) as a function of the WSe\(_2\) gate voltage \( V_g \) for the local and non-local CPGE results. A monotonic increase is observed, similar to the case of local WSe\(_2\) CPGE measurements.
shown that the inversion-symmetry breaking plays a major role in generating the valley-polarized DoF with spin DoF textures partially governed by Rashba spin splitting. The linear dependence of $C$ on $V_g$ (Fig. 2a) can be understood through the following equation:\(^\text{14}\)

\[ C_{\text{valley}} \sin 2\phi = \chi gE_{\text{ex}}|M^{i\rightarrow f}| \sin \theta \sin 2\phi \]  

(3)

where $M$ is the transition amplitude from the conduction band $i$ (initial state) to $f$ (final state) at the $\Lambda$ ($\Lambda'$) valley, $\chi$ is the coefficient for the band $f$, $l$ is the incident light intensity, $g$ is the coefficient associated with the time-reversal and reflection symmetries. Equation (3) implies that the local CPGE $C$ in principle should exhibit a superlinear dependence on the applied electric field $E_{\text{ex}}$. This is because increasing $V_g > 0$ not only enhances the inversion-symmetry breaking, but also induces more electron doping on the top surface of WSe$_2$. However, the observed monotonic increase of $C$ deviates somewhat from this expectation; it may originate from the additional contributions of linear photogalvanic $L_{\Delta}$ and $L_{\Gamma}$ that can also be enhanced by the doping effect.\(^{15,16}\) The local $P$ (Fig. 3d), on the other hand, is invariant under such doping effects; in equation (2), we see that both the numerator $C$ and denominator $C$ and $L$ scale equally with $V_g$. Thus, the $V_g$-induced inversion-symmetry breaking leads to the linear $V_g$ dependence for $P$. The observed nonzero $P$ when $V_g = 0$V indicates the presence of an internal built-in electric field at the interface between the ionic liquid and top layer of WSe$_2$.\(^{14,27}\)

A close examination of the valley and spin DoF formalism shows why our integrated lateral heterostructure is unique for selective spin DoF detection. The valley-locked spin DoF $C_{\text{valley}, \text{spin}}$ and the valley DoF $C_{\text{valley}}$ in the local WSe$_2$ CPGE have the following relationships:\(^{14,18}\) (Supplementary Information III):

\[ C_{\text{valley}, \text{spin}} \sin 2\phi \approx (\beta^d - \beta^i)C_{\text{valley}} \sin 2\phi \]  

(4)

where $\beta$ is the second-rank pseudo-tensor of anti-symmetric split for initial ($i$) and final ($f$) states, which describes the control of spin polarization by an externally applied electric field. Because the photocurrent direction and the helicity-dependent optical excitation are mathematically the same for both valley and spin DoF carriers, it is impossible to distinguish the two DoFs using the WSe$_2$ transistor alone. On the other hand, the integrated TMD–graphene–TI device enables selective spin DoF transport. In our device, the valley DoF cannot be all transferred to TI, because of the large momentum mismatch between the TMD and TI and the extremely fast valley depolarization.\(^{28–30}\)

The presented analysis demonstrates that our hetero-device acts as a platform to extract the pure spin DoF photocurrent from strongly coupled valley-spin DoF carriers via non-local detection. Since the non-local CPGE response is locked with the valley-polarized current in WSe$_2$ TMD ($C_{\text{valley}}$),\(^{14}\) the non-local $V_g$-dependent $P$ shown in Fig. 3e reflects the local $P$ response of Fig. 3d. In addition to this linear dependence, because the Rashba spin splitting ($\beta^d - \beta^i$) in equation (4) also includes the $V_g$-dependent term, the non-local $P$ (Fig. 3e) exhibits a quadratic $V_g$ dependence.

In conclusion, we demonstrated room-temperature optoelectronic transport of the valley-locked spin polarization DoF using WSe$_2$–graphene–Bi$_2$Se$_3$ lateral heterostructures. The optically generated valley–spin coupled DoFs in WSe$_2$ diffuse across the monolayer graphene, and non-local electrical transport selectively measures the spin DoF at the Bi$_2$Se$_3$ TI electrodes. Electrical tunability was demonstrated, where the non-local quadratic increase of $P$ with $V_g$ is in remarkable contrast to the local linear increase of $P$. For potential future applications, although our heterostructure shows $P$ as high as 55%, the spin DoF injection efficiency is still low. The spin DoF depolarization may be circumvented by employing a hot-electron ballistic tunnel contact\(^{11}\) using atomically thin hexagonal boron nitride (h-BN)\(^{32}\) at each lateral junction interface. Our device platform for manipulating the valley and spin DoFs is expected to pave the way towards future spintronic and valleytronic applications.

Methods
Methods, including statements of data availability and any associated accession codes and references, are available at https://doi.org/10.1038/s41565-018-0195-y.
References


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Author contributions

S.C. and M.N. contributed equally to this work. H.C. conceived the main idea and supervised the project. S.C., M.N. and H.C. wrote the manuscript with input from all authors.

Competing interests

The authors declare no competing interests.

Additional information

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Methods

Device preparation. WSe$_2$, monolayer graphene, and Bi$_2$Se$_3$ were obtained by mechanical exfoliation from bulk crystals. The WSe$_2$ crystal and graphite were bought from HQ Graphene, and the single crystal of Bi$_2$Se$_3$ was grown using the melting method. The stoichiometric mixture of Bi (99.999%) and Se (99.9999%) was loaded in an evacuated quartz ampoule, which was heated at 850 °C for 48 h, followed by slow cooling to 500 °C at a rate of 2 °C per hour and post-annealing at 500 °C for a further five days before furnace cooling. The topological surface state of Bi$_2$Se$_3$ crystals was confirmed by the angle-resolved photoemission spectroscopy, as shown in ref. 33. Using a polydimethylsiloxane (PDMS) viscoelastic stamping technique, the WSe$_2$ and Bi$_2$Se$_3$ flakes were transferred sequentially onto a Si/SiO$_2$ substrate while minimizing the distance between them. At the edge of the Bi$_2$Se$_3$ TI flake, we applied electron-beam-overexposed PMMA to minimize the contribution from the spin-insensitive bulk conduction. Electrodes were deposited by standard electron beam lithography with Ti (20 nm) and Au (110 nm) layers to serve as contacts for the electrical and CPGE measurements. Then, monolayer graphene was transferred and placed between the WSe$_2$ and Bi$_2$Se$_3$ flakes with a PMMA-based wet transfer process. Before the graphene transfer, we removed the naturally grown oxide layer from the surface of the Bi$_2$Se$_3$ flake using reactive-ion etching (Ar 25 sccm, 30 s). Finally, ionic liquid N,N-diethyl-N-methyl-N-(2-methoxyethyl) ammoniumbis (trifluoromethylsulphonyl) imide (DEME-TFSI) was drop-cast onto the device (Supplementary Information I for the fabrication procedure and Supplementary Information V for the energy band diagram).

Data availability. The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

References