Supplementary information to:
Symmetry regimes for circular photocurrents in monolayer MoSe₂

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Supplementary Note 1: AFM characterization

We measure the height profile of the BN-encapsulated MoSe$_2$ on a SiO$_2$/Si substrate by AFM. The thickness of both of the MoSe$_2$ and the top h-BN flakes are measured as 0.7 nm (Supplementary Figure 1b) which corresponds to monolayer MoSe$_2$ and bilayer h-BN, in agreement with the reported values in literature$^{1,2}$. The bottom h-BN has a thickness of 7.65 nm (21-22 layers). The AFM images also reveal the presence of bubbles due to trapped molecules in the h-BN/MoSe$_2$ interface. Reportedly, the accumulation of the interface contaminants in these bubbles ensures a perfectly clean interface at the bubble-free regions, and is a signature of the good adhesion between the two layers.$^3$

Supplementary Figure 1 (a) AFM image of the BN-encapsulated MoSe$_2$ on SiO$_2$/Si substrate. The dashed lines highlight the edge of the flakes. (b) Height profile along the red and blue lines indicated in panel (a), corresponding to the edges of the monolayer MoSe$_2$ and the bilayer h-BN flakes. For clarity, the profiles are offsetted to the same zero level.
**Supplementary Note 2:** Optical microscopy images of the fabrication process and the final device

![Image](image_url)

**Supplementary Figure 2** Enhanced-contrast optical images of the device fabrication process (a) Fully encapsulated 1L-MoSe\(_2\) crystal before the fabrication of the contacts. (b) Final device. (c-e) Zoom in of the region indicated by a dotted square in panels (a) and (b) at the different stages of the fabrication process. (c) Exfoliated 1L-MoSe\(_2\) flake on SiO\(_2\) before processing. (d) Same flake shown in (c) after encapsulation with top bilayer h-BN and bottom multilayer h-BN. (e) Final device with the fabricated contacts on top of the BN/MoS\(_2\)/BN stack. The electrodes used for the measurements of the main text are highlighted in green. The dashed white line indicates the edges of the 1L-MoSe\(_2\) flake.
Supplementary Note 3: Electrical characterization of the 1L-MoSe₂ phototransistors

The DC electrical characterization of the sample is performed in the dark while keeping the sample in vacuum (10⁻⁴ mbar). In order to obtain the electrical transport properties of the MoSe₂ channel, we perform four-terminal measurements in Hall-bar geometry. We apply a source-drain current on the contacts 1 and 2 (See figure 1a in the main text) and measure the voltage drop along the channel using the Hall contacts A and C. We remark that using the contacts that only partially cover the channel is preferable for the characterization of the intrinsic electrical properties of the MoSe₂ channel, since this allows to prevent the formation of depletion regions near the metal contacts.⁴

Supplementary Figure 3a shows a transfer characteristic for the 1L-MoSe₂ phototransistor, presenting a clear n-type behavior. We extract the threshold gate voltage ($V_{th}$) of 19 V as the gate voltage at which the conductivity starts to increase. We estimate a field-effect mobility of about 17 cm²/V.s from the linear fit to the transfer curve, for the range of gate voltage ($V_{gate} > V_{th}$) with linear dependence of conductivity. Supplementary Figure 3b shows the four-terminal I-V characteristics of the phototransistor. The ohmic response of the channel can be readily observed from the linearity of the obtained I-Vs. The inset in Supplementary Figure 3b shows the square resistance of the MoSe₂ channel, $R_{sq}$, obtained as the slope of the linear fit to the I-V divided by the length-to-width ratio of the MoSe₂ channel, as a function of the gate voltage.

In our device geometry, encapsulation of the MoSe₂ channel with h-BN reduces the influence of the adsorbate molecules on the MoSe₂ surface and prevents charge scatterings due to interface impurities and the Si substrate, which largely reduces the hysteresis in the charge transport measurements. Moreover, the bilayer h-BN plays the role of a tunnel barrier for injection of charge carriers, preventing the level pinning at the contacts.

Supplementary Figure 3  Electrical characterization of the channel in 4-terminal geometry. (a) Channel conductivity as a function of gate voltage. The red line is a linear fit to the data for $V_{gate} > V_{th}$. (b) I-V characteristics of the channel and the square resistances, estimated for the gate voltages of 40 to 60 V (shown in the legend).
**Supplementary Note 4:** Color map of the CPC amplitude as a function of $V_{ds}$ and $V_{gate}$ for illumination at normal incidence.

Supplementary Figure 4 shows a colormap of the CPC amplitude $C$ as a function of the drain-source and gate voltages for $\phi = 0^\circ$. The value of $C$ remains near zero regardless of the applied voltages. This allows us to rule out that the dominant contribution to our observed CPC signals is a Berry phase-induced CPGE, since it should become maximal for normal incidence. This measurement also rules out that our signals have a significant contribution from the valley-Hall effect, since such effect would appear as a nonzero contribution to the CPC linear with the drain-source voltage. The absence of the valley-Hall effect in our device can be understood since this effect has been reported for studies on highly n-doped devices, and it increases with the gate voltage. In our device, the 1L-MoSe$_2$ channel only starts to open for $V_{gate} > 20$ V. Thus, a much larger doping could be required for observing the valley-Hall effect.
**Supplementary Note 5: Comparison of the photovoltage and photocurrent measurements and consistency checks**

Supplementary Figure 5 shows the helicity-dependent open-circuit photovoltage and short-circuit photocurrent measured in two sets of electrodes: [A, B] and [1, 2]. These results are representative for a wider range of checks that we performed, where we always found a linear relation between the observed values for $C$, $L_1$ and $L_2$ in the current and voltage signals. This photoresponse can thus be measured equivalently as current or voltage signals on our device. In addition, for $C$ we observed no dependence on the orientation of the linear polarization for the laser beam incident on the $\lambda/4$ plate.

Finally, Figs. 2b and 3c (main text) show that the spectral dependence of $C$ is preserved for two different sets of electrodes, further ruling out a role for specific contacts or standing-wave effects between electrodes. Our full range of consistency checks allows us to rule out that effects at specific electrodes, and effects from confining light between the micron-scale metallic electrode structure, give a significant contribution to the helicity-dependent signals that we analyze.

**Supplementary Figure 5** Helicity-dependent open-circuit photovoltage (red circles) and short-circuit photocurrent (blue circles) for electrodes [1, 2] (a) and [A, B] (b) at $V_{gate} = 0$ V and $V_{ds} = 0$, as a function of the waveplate angle ($\theta$). The black solid lines are fittings to the phenomenological equation (1) in the main text. Except for a scale factor, the $\theta$ dependence of the photovoltage and photocurrent are very similar, as expected from the linear I-V of the semiconductor channel.
Supplementary Figure 6 shows the power dependence of C, L₁ and L₂ for electrodes A and B and measuring photocurrent instead of photovoltage, in contrast with the measurements shown in the main text for contacts 1 and 2 (Figure 1c). This shows that the same linear power dependence appears regardless of the contacts or the measurement technique.

Supplementary Figure 6: Illumination power dependence of L₁, L₂ and C extracted from the fittings of helicity-dependent photocurrent measurements (as those shown in Supplementary Figure 5) for contacts A and B.
Supplementary Note 6: Theoretical analysis of the photogalvanic and photon drag effects

I. General description of photogalvanic and photon drag effects

The theoretical foundations of the photogalvanic and photon drag effects (PGE and PDE) trace back to several decades ago. More specifically, they are characterized by a DC current generated by a time-varying electric field, with amplitude proportional to the square of the applied field. This current is generated by photoelectrons which are excited by optical (vertical in the band structure) transitions and, depending on its microscopic origin, can depend on the polarization (linear PCE and PDE) or the helicity (circular PGE and PDE) of the applied field. Recent studies on derivations of PGE and PDE rely on the nonlinear susceptibility, Floquet theory and the kinetic equation approach.

Let us consider the situation of a 2D material illuminated by a monochromatic light source, with complex electric field defined as the plane wave

$$E_j(\vec{r}, t) = E_je^{i\omega t+ i\vec{q} \cdot \vec{r}} + E_j^*e^{i\omega t- i\vec{q} \cdot \vec{r}},$$

where the subindices $i, j$ and $k$ stand for the Cartesian coordinates, $\omega$ is the angular frequency and the wave vector $\vec{q}$ can be expressed in spherical coordinates as

$$\vec{q} = -q(\sin(\phi) \cos(\alpha), \sin(\phi) \sin(\alpha), \cos(\phi)).$$

We assume that the incident light forms a polar angle $\phi$ and an azimuthal angle $\alpha$ with the 2D plane (see Figure 1 in the main text). We can write down the electric field (as well as the vector potential $\vec{A} = \vec{E}/i\omega$) as

$$\vec{E} = \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} = E_0 \begin{pmatrix} -i \sin(2\theta) \cos(\alpha) + (1 - i \cos(2\theta)) \cos(\phi) \cos(\alpha) \\ i \sin(2\theta) \cos(\alpha) + (1 - i \cos(2\theta)) \cos(\phi) \sin(\alpha) \\ -(1 - i \cos(2\theta)) \sin(\phi) \end{pmatrix},$$

with $E_0$ as the magnitude of the applied electric field and $\theta$ as the angle between the fast axis of the $\lambda/4$ waveplate and the initial linear polarization of the light. $\theta = \pi/4, (3\pi/4)$ for left (right) circularly polarized light. For future purpose, here we write $(\vec{E} \times \vec{E}^*)$ as

$$\begin{align*}
(\vec{E} \times \vec{E}^*)_x &= -2i \cos(\alpha) \sin(2\theta) \sin(\phi) E_0^2 \\
(\vec{E} \times \vec{E}^*)_y &= -2i \sin(\alpha) \sin(2\theta) \sin(\phi) E_0^2 \\
(\vec{E} \times \vec{E}^*)_z &= -2i \sin(2\theta) \cos(\phi) E_0^2
\end{align*}$$

The light-induced current density $\vec{J}$ inside the material can be generically written in series of the Cartesian components $(l, j, k)$ of the electric field $\vec{E}$. Per component of $\vec{J}$ this gives

$$J_l = \sigma_{lj}E_l e^{-i\omega t+ i\vec{q} \cdot \vec{r}} + \sigma_{ljk}^{(2)} E_l E_k e^{-2i\omega t+ 2i\vec{q} \cdot \vec{r}} + \sigma_{ljk}^{(3)} E_l E_k E_l + \cdots,$$

where $\sigma_{lj}$ is a second rank tensor and $\sigma_{ljk}^{(2)}$, and $\sigma_{ljk}^{(3)}$ are third rank tensors. The first and second terms in the right correspond respectively to a linear AC current (at optical frequency) in response to the electric field and an AC
current of twice the frequency of the radiation, responsible for second harmonic generation. The relevant term for us is the third term, which corresponds to a DC current \( J_{i}^{\text{DC}} \) in response to the oscillating field:

\[
J_{i}^{\text{DC}} = \sigma_{ijk}^{(2)} E_{j} E_{k}^*,
\]

By doing a Taylor expansion over the wave vector \( q \) we can rewrite \( J_{i}^{\text{DC}} \) as

\[
J_{i}^{\text{DC}} = \sigma_{ijk}^{(2)}(\omega, \vec{q}) E_{j} E_{k}^* = \chi_{ijk}(\omega) E_{j} E_{k}^* + T_{ijk\mu}(\omega) q_{\mu} E_{j} E_{k}^* + \cdots
\]

Here \( \chi_{ijk} = \sigma_{ijk}^{(2)}(\omega, 0) \) does not depend on the radiation wave vector \( \vec{q} \) and is responsible for the photogalvanic effect, \( J_{i}^{\text{PGE}} \), while \( T_{ijk\mu} \) accounts for the photon drag effect \( J_{i}^{\text{PDE}} \), linear with factors \( q_{\mu} \).

**Requirement of inversion symmetry breaking**

We now show that the absence of inversion symmetry is necessary for getting nonzero photogalvanic and photon drag effect. First, we note that \( J_{i}^{\text{DC}} \) is antisymmetric (changes its sign) under inversion of the spatial coordinates \( x, y, z \rightarrow -x, -y, -z \), while the object \( E_{j} E_{k}^* \) is symmetric under that transformation. In consequence, if the inversion transformation is a symmetry of the studied system, \( J_{i}^{\text{DC}} \) cannot have any dependence on \( E_{j} E_{k}^* \) and \( \chi_{ijk} \) must be zero. In other words, the photogalvanic effect can only emerge in systems with broken inversion symmetry.

**Linear and circular photogalvanic and photon drag effect**

Next, we observe that, since the current density must be real, it cannot change under complex conjugation. In consequence, from equation (7) we get \( \chi_{ikj} = \chi_{ijk} \). Therefore, the real part of \( \chi_{ijk} \) is symmetric under coordinate exchange while its imaginary part is antisymmetric under this operation. This allows us to rewrite the photogalvanic current as follows,

\[
J_{i}^{\text{PGE}} = \chi_{ijk} E_{\beta} E_{\gamma}^* = \chi_{ijk}^{\text{sym}} E_{j} E_{k}^* + i \chi_{ijk}^{\text{antisym}} E_{j} E_{k}^* ,
\]

or, using the transformation under permutation of the subindices \( j \) and \( k \),

\[
J_{i}^{\text{PGE}} = \frac{1}{2} \chi_{ijk}^{\text{sym}} (E_{j} E_{k}^* + E_{k} E_{j}^*) + \frac{1}{2} \chi_{ijk}^{\text{antisym}} (E_{j} E_{k}^* - E_{k} E_{j}^*) \equiv J_{i}^{\text{LPGE}} + J_{i}^{\text{CPGE}}
\]

We can now compare \( J_{i}^{\text{LPGE}} \) and \( J_{i}^{\text{CPGE}} \) with the Stokes parameters:

\[
S_{1} = \frac{|E_{x}|^2 - |E_{y}|^2}{|E_{x}|^2 + |E_{y}|^2}; \quad S_{2} = \frac{E_{x} E_{y}^* + E_{y} E_{x}^*}{|E_{x}|^2 + |E_{y}|^2}; \quad S_{3} = i \frac{E_{x} E_{y}^* - E_{y} E_{x}^*}{|E_{x}|^2 + |E_{y}|^2}
\]

We see that \( J_{i}^{\text{LPGE}} \) is proportional to \( S_{2} \), which accounts for the linearly polarized radiation (linear photogalvanic effect), while \( J_{i}^{\text{CPGE}} \) is proportional to \( S_{3} \), and, therefore, it is sensitive to the circularly polarized radiation (circular photogalvanic effect).
Finally, the totally antisymmetric Levi-Civita tensor $\epsilon_{sjk}$ can be used to contract $\chi^\text{antisym}_{ijk}$ to only one pseudo vector index,

$$\sum_{jk} \chi^\text{antisym}_{ijk} (E_jE_k^* - E_kE_j^*) = i \sum_{sjk} 2\gamma_{ls} \epsilon_{sjk} (E_jE_k^* - E_kE_j^*) = i \sum_s \gamma_{ls} (\vec{E} \times \vec{E}^*) \cdot \epsilon_{s},$$

(11)

where $\gamma_{ls}$ is a second rank pseudo-tensor and $l$ and $s$ stand for Cartesian coordinates. Thus $J_l^{\text{CPGE}}$ can be expressed as

$$J_l^{\text{CPGE}} = i \sum_j \gamma_{lj} (\vec{E} \times \vec{E}^*)_j ,$$

(12)

It is convenient to separate $J_l^{PDE}$ in a similar fashion into its circular and linear polarization sensitive components, $J_l^{\text{CPDE}}$ and $J_l^{\text{LPDE}}$. For $J_l^{\text{CPDE}}$ we get:

$$J_l^{\text{CPDE}} = i \sum_{jk} T_{ljk} q_j (\vec{E} \times \vec{E}^*)_k ,$$

(13)

At this point it is worth noting that we have not still made any assumption about the physical origin of $J_l^{\text{CPGE}}$. Thus, equations (12) and (13) are completely general and must hold regardless of the underlying physical mechanism.

II. Symmetry arguments for the CPC in monolayer TMDCs.

In the following, we use symmetry arguments to determine the nonzero components of the tensors $\gamma_{ij}$ and $T_{lsj}$, as defined in equations (12) and (13). This allows to extract constraints for the dependence of $J_l^{\text{CPGE}}$ and $J_l^{\text{CPDE}}$ on the illumination angles $\alpha$ and $\phi$. We remark again that, since equations (12) and (13) must hold regardless of the physical origin of the CPC, the discussion below is completely general.

In order of decreasing symmetry we analyze three cases: $D_{3h}$, $C_{3v}$, and Single mirror-plane symmetry. We find that crystal structures belonging to the high-symmetry class $D_{3h}$ cannot support any CPGE. For the case of $C_{3v}$ symmetry with an oblique incidence angle $\phi$, only $\gamma_{xy}$ can have a nonzero value, which then gives a nonzero CPGE, but always with the property that it flips signs upon reversal of $\phi$ (in conflict with a BC origin). Systems with only one mirror symmetry can not only have nonzero $\gamma_{xy}$ but also nonzero $\gamma_{yz}$ and $\gamma_{xz}$, allowing for BC-CPGE. We conclude that our experimental results for low source-drain voltage are only compatible with, at most, one mirror-plane symmetry, since otherwise $\gamma_{xz}$ and $\gamma_{yz}$ cancel out and, therefore, the photocurrent cannot be preserved upon inversion of the incidence angle, $\phi$. 


**D₃h symmetry**

The scenario of a 1L-TMDC system with ideal mirror symmetry with respect to the crystal plane (symmetric environments and external fields) gives the system the high $D_{3h}$ symmetry. This has a three-fold rotation symmetry around the $z$ axis, defined by the operator $C_3$, three two-fold axes perpendicular to $C_3$, a mirror plane in the $xy$ plane, defined by $\sigma_h = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{pmatrix}$ and three mirror vertical planes with respect to the $xy$ plane $\sigma_v$. The improper rotation is $\sigma_hC_3$.

In the section directly below here on $C_{3v}$ symmetry we derive that the CPGE current is

$$j_{CPGE}^{C_{3v}} = i \begin{pmatrix} 0 \\ -y_{xy} \\ 0 \end{pmatrix} \begin{pmatrix} (\vec{E} \times \vec{E}^*)_x \\ (\vec{E} \times \vec{E}^*)_y \\ (\vec{E} \times \vec{E}^*)_z \end{pmatrix} = i \begin{pmatrix} y_{xy}(\vec{E} \times \vec{E}^*)_y \\ -y_{xy}(\vec{E} \times \vec{E}^*)_x \\ 0 \end{pmatrix}. \quad (14)$$

This result for $C_{3v}$ can be extended to the case for $D_{3h}$ by adding the requirement for the additional mirror symmetry $\sigma_h$. This brings that a pseudo-vector $(\vec{E} \times \vec{E}^*)$ becomes $-\sigma_h(\vec{E} \times \vec{E}^*)$, and gives the condition $y_{xy} = 0$. Consequently, all CPGE current contributions cancels out for the $D_{3h}$ symmetry.

For CPDE, the $C_{3v}$ symmetry (below) yields

$$j_{CPDE}^{C_{3v}} = i (T_{yxz} + T_{yzx})q_e \begin{pmatrix} -(\vec{E} \times \vec{E}^*)_y \\ (\vec{E} \times \vec{E}^*)_x \\ 0 \end{pmatrix} \propto \begin{pmatrix} -\sin(\alpha) \sin(2\theta) \sin(2\phi) \\ \cos(\alpha) \sin(2\theta) \sin(2\phi) \\ 0 \end{pmatrix}. \quad (15)$$

A direct extension of this analysis shows that the additional mirror plane $\sigma_h$ does not impose further constrains on $j_{CPDE}^{C_{3h}}$. Thus, CPDE photocurrents can be present for this symmetry.

**C₃v symmetry**

If we assume a 1L-TMDC crystal symmetry in the plane, but drop the assumption of mirror symmetry with respect to the crystal plane (relevant, for example, for a 1L-TMDC sustained on a substrate), the system has $C_{3v}$ symmetry. This corresponds to a three-fold rotation symmetry around the $z$ axis $C_3$, and three mirror planes perpendicular to the $xy$ plane. The CPGE photocurrent is given by

$$j_{iCPGE}^{C_{3v}} = i \gamma_{ij}(\vec{E} \times \vec{E}^*)_j. \quad (16)$$

Under a $2\pi/3$ rotation

$$R = \begin{pmatrix} \cos(2\pi/3) & \sin(2\pi/3) & 0 \\ -\sin(2\pi/3) & \cos(2\pi/3) & 0 \\ 0 & 0 & 1 \end{pmatrix},$$

$\vec{E} \times \vec{E}^*$ becomes $R(\vec{E} \times \vec{E}^*)$ and $j_{CPGE}^{C_{3v}}$ becomes $Rj_{CPGE}^{C_{3v}}$. Since $\gamma$ should remain the same under the rotational symmetry, we have
\[ R\mathcal{J}^{\text{CPGE}} = i\gamma R(\vec{E} \times \vec{E}^*) . \]  

(17)

By replacing (26) into (27) we obtain
\[ R\gamma = \gamma R . \]  

(18)

An additional constraint is given by the mirror symmetry. There are three mirror planes perpendicular to the \( xy \) plane, and we assume that the angle between the mirror plane and the \( x \) axis is \( \psi \). Under the mirror reflection, characterized by the operator
\[ \begin{pmatrix}
\cos(2\psi) & \sin(2\psi) & 0 \\
\sin(2\psi) & -\cos(2\psi) & 0 \\
0 & 0 & 1
\end{pmatrix} , \]
\( \gamma \) should remain the same, while \( \mathcal{J}^{\text{CPGE}} \) becomes \( M\mathcal{J}^{\text{CPGE}} \). As a pseudo-vector, \((\vec{E} \times \vec{E}^*)\) becomes \(-M(\vec{E} \times \vec{E}^*)\). Therefore, we obtain
\[ M\gamma = -\gamma M . \]  

(19)

Combining the constraints from rotational and mirror symmetries, we conclude that \( \gamma_{ij} \) has only one independent parameter:
\[ \gamma_{ij} = \begin{pmatrix}
0 & \gamma_{xy} & 0 \\
-\gamma_{xy} & 0 & 0 \\
0 & 0 & 0
\end{pmatrix} , \]  

(20)

and the CPGE current must have a form of
\[ \mathcal{J}^{\text{CPGE}} = i \begin{pmatrix}
0 & \gamma_{xy} & 0 \\
-\gamma_{xy} & 0 & 0 \\
0 & 0 & 0
\end{pmatrix} \begin{pmatrix}
(\vec{E} \times \vec{E}^*)_x \\
(\vec{E} \times \vec{E}^*)_y \\
(\vec{E} \times \vec{E}^*)_z
\end{pmatrix} \propto \gamma_{xy} \begin{pmatrix}
\sin(\alpha) \sin(2\theta) \sin(\phi) \\
-\cos(\alpha) \sin(2\theta) \sin(\phi) \\
0
\end{pmatrix} . \]  

(21)

This description yields a nonzero CPGE current that indeed shows a \( \sin(2\theta) \) dependence in the polarization control. Further, it only yields nonzero CPGE currents for nonzero angles \( \phi \). For this symmetry group the CPGE current changes sign when the incidence angle is switched from \( \phi \) to \(-\phi \).

For the CPDE, we have
\[ J_{i}^{\text{CPDE}} = iT_{ijk}q_{j}(\vec{E} \times \vec{E}^*_k) . \]

Impose that, for a transverse electromagnetic wave, the vector \((\vec{E} \times \vec{E}^*_k)\) should be along the same direction as the photon momentum \( \vec{q} \), we get \( q_{j}(\vec{E} \times \vec{E}^*_k)_k = q_{k}(\vec{E} \times \vec{E}^*_j)_j \).

Impose the invariance of \( \mathcal{J}^{\text{CPDE}} \) under a rotation of \( 2\pi / 3 \), \( R\mathcal{J}^{\text{CPDE}} = R\mathcal{J}^{\text{CPDE}} \) we get
\[ T_{zil}R_{ik}q_{k}R_{jl}(\vec{E} \times \vec{E}^*_i)_l = T_{zij}q_{l}(\vec{E} \times \vec{E}^*_i)_j . \]  

(22)

From each \( q_{l}(\vec{E} \times \vec{E}^*_i)_j \) we get
\[ T_{xx} + T_{zx} = T_{zy} + T_{xz} = T_{zz} + T_{zx} = 0 ; \]
\[ T_{zy} = T_{zx} . \]  

(23)

Also, from \( j_z \) and \( j_y \) we get
\[
\begin{align*}
T_{xxz} &= 0; \quad T_{xyy} = -T_{xxx}; \quad T_{yxx} = \sqrt{3}T_{xxx}; \quad T_{xy} + T_{yy} = -2T_{xxx}; \\
T_{yxz} + T_{yzx} &= -(T_{xy} + T_{xx}); \quad T_{yy} + T_{yz} = T_{xx} + T_{xxx}; \quad T_{yy} = \frac{1}{2}(T_{xy} + T_{xx}). 
\end{align*}
\] (24)

Combining this with the restrictions imposed by the mirror symmetry we find that only \( T_{yxx} + T_{yzx} = -\left( T_{xy} + T_{xx} \right) \equiv \chi \) remains and

\[
\tilde{j}_{CPDE}^{CPDE} = i(T_{yxx} + T_{yzx})q_z \begin{pmatrix}
-\left( \vec{E} \times \vec{E}^* \right)_y \\
\left( \vec{E} \times \vec{E}^* \right)_x \\
0
\end{pmatrix} \propto \begin{pmatrix}
-\sin\alpha \sin(2\theta) \sin(2\phi) \\
\cos\alpha \sin(2\theta) \sin(2\phi) \\
0
\end{pmatrix}.
\] (25)

Remarkably, under \( C_3 \) symmetry we find that a CPGE must have a \( \sin(\phi) \) dependence, while a CPDE must have a \( \sin(2\phi) \) dependence. They can thus be distinguished by their dependence on \( \phi \).

**Single mirror-plane symmetry**

Now we consider a case of even lower symmetry: that of a 1L-TMD system that has just one mirror plane which is perpendicular to the \( xy \) plane. In a real device, the presence of asymmetric electrodes and strain gradients is expected to lead to this low-symmetry situation (or even lower symmetry). In particular, this scenario is relevant for strained monolayer MoSe₂, since the lowest stiffness for deformation occurs along the armchair direction in the crystal. We cannot assume that this crystal direction has a known relation with the \( xy \) coordinate frame (defined by the experimental geometry with electrodes, see Fig. 1 main text). We therefore introduce the angle \( \psi \) to describe angle between the mirror plane and the \( x \) axis.

Under the mirror reflection, \( j_{CPGE}^{CPGE} \) becomes \( Mj_{CPGE}^{CPGE} \), where \( M = \begin{pmatrix}
\cos(2\psi) & \sin(2\psi) & 0 \\
\sin(2\psi) & -\cos(2\psi) & 0 \\
0 & 0 & 1
\end{pmatrix} \). Once more, as a pseudovector, \( \left( \vec{E} \times \vec{E}^* \right) \) becomes \( -M\left( \vec{E} \times \vec{E}^* \right) \). Therefore, we obtain \( M\gamma = -\gamma M \). The absence of the \( 2\pi/3 \) rotational symmetry in this case allows more independent parameters to appear in \( \gamma_{ij} \) and the CPGE current takes the form

\[
\tilde{j}_{CPGE} = i \begin{pmatrix}
0 & \gamma_{xy} & \gamma_{xz} \\
\gamma_{xy} & 0 & \gamma_{yz} \\
\gamma_{xz} & \gamma_{yz} & 0
\end{pmatrix} \begin{pmatrix}
\left( \vec{E} \times \vec{E}^* \right)_x \\
\left( \vec{E} \times \vec{E}^* \right)_y \\
\left( \vec{E} \times \vec{E}^* \right)_z
\end{pmatrix} \propto \begin{pmatrix}
\gamma_{xy}(\sin\alpha \sin(\phi) + \gamma_{xx} \cos(\phi)) \sin(2\theta) \\
\gamma_{xy}(\cos\alpha \sin(\phi) + \gamma_{xx} \cos(\phi)) \sin(2\theta) \\
0
\end{pmatrix},
\] (26)

where we assumed that the \( j_{z}^{CPGE} \) component must be zero for a single-layer crystal.

The mirror symmetry yields the additional conditions:

\[
\gamma_{xz} = \frac{1 - \cos(2\psi)}{\sin(2\psi)} \gamma_{yz} \quad \text{and} \quad \gamma_{zx} = \frac{1 - \cos(2\psi)}{\sin(2\psi)} \gamma_{zy}.
\] (27)

(28)
Thus, we get a CPGE contribution independent of \( \alpha \) and changing as \( \cos(\phi) \). In the next section we will show that this angular dependence is required for a Berry curvature-induced CPGE.

Now we consider the CPDE. For simplicity, we assume that the mirror symmetry is from \( x \) to \( -x \). We get

\[
\begin{align*}
    j_x^{\text{CPDE}} &= 2 \sin(2\theta) \left\{ T_{xzz} \cos^2(\phi) + \frac{T_{xyz} + T_{xzy}}{2} \sin(\alpha) \sin(2\phi) \\
    &\quad + (T_{xxx} \cos^2(\alpha) + T_{xxy} \sin^2(\phi)) \sin^2(\phi) \right\}; \\
    j_y^{\text{CPDE}} &= \sin(2\theta) \left\{ (T_{yxz} + T_{xyz}) \cos(\alpha) \sin(2\phi) + (T_{yxy} + T_{yyx}) \sin(2\alpha) \sin^2(\phi) \right\}; \\
    j_z^{\text{CPDE}} &= \sin(2\theta) \left\{ (T_{zzx} + T_{zxx}) \cos(\alpha) \sin(2\phi) + (T_{zzy} + T_{zyx}) \sin(2\alpha) \sin^2(\phi) \right\};
\end{align*}
\]

Here, we find that CPDE can have a contribution dependent on \( \sin(2\alpha) \sin^2(\phi) \), which matches the observed angular dependence for the low-\( V_{ds} \) regime.

As discussed in the main text, this symmetry analysis does not bring forward the \( \sin(3\alpha) \) dependence experimentally observed for the large-\( V_{ds} \) regime (although this could still come forward from asymmetric transport properties of the zig-zag and armchair directions of the MoSe\(_2\) crystal).

In the particular case where an additional mirror symmetry in the \( z \) direction is allowed (original \( D_{3h} \) case subject to e.g. uniaxial strain), the CPGE and CPDE current is further constrained, yielding

\[
\begin{align*}
    \mathbf{j}^{\text{CPGE}} &= i \begin{pmatrix} 0 & 0 & y_{xz} \\ 0 & 0 & y_{yz} \\ y_{zx} & y_{zy} & 0 \end{pmatrix} \begin{pmatrix} \vec{E} \times \vec{E}^* \end{pmatrix}_x \propto \begin{pmatrix} y_{xz} \cos(\phi) \sin(2\theta) \\ y_{yz} \cos(\phi) \sin(2\theta) \\ 0 \end{pmatrix}, \\
    \mathbf{j}^{\text{CPDE}} &= \begin{pmatrix} (T_{xyz} + T_{xyz}) \sin(\alpha) \sin(2\phi) \sin(2\theta) \\ (T_{yxz} + T_{xyz}) \cos(\alpha) \sin(2\phi) \sin(2\theta) \\ (T_{zyx} + T_{zxx}) \cos(\alpha) \sin(2\phi) \sin(2\theta) \end{pmatrix}.
\end{align*}
\]

and only the \( \cos(\phi) \)-dependent CPGE contribution can still appear. Also, for the CPDE current, the terms on \( \sin(2\alpha) \sin^2(\phi) \) fade out. Thus, we see that our experimental results require a broken out-of-plane mirror symmetry.

III. Berry curvature and circular photogalvanic effect

In this section, we show how a circular photogalvanic current can emerge as a consequence of a nonzero Berry curvature. We start the derivation of the photocurrent equation based on the assumption that the momentum of light is small and can be ignored. Thus, we consider only vertical optical interband transition between the initial and final bands. The photocurrent \( \mathbf{j} \) can be derived based on the Fermi-Golden rule \( ^{10} \):

\[
\begin{align*}
    J_i &= -\frac{2\pi e}{h} \sum_{l,F} \frac{d^2k}{(2\pi)^2} f_{fi}(k) (v_{fl}^i - v_{fl}) \delta (\Delta E_{fi} - \omega) |D|^2.
\end{align*}
\]
where \( v \) is the group velocity of the electron state, \( \omega \) is the excitation energy, \( \Delta E_{Fi} = E_F(k) - E_i(k) \) and \( f_{1F}(k) \equiv f_i(k) - f_F(k) \) are the differences of energy and equilibrium Fermi distribution function between the initial and final states. \( |i⟩ \) and \( |F⟩ \) are the Bloch wavefunction of the initial and final states. \( D \) is the optical transition dipole defined as

\[
D = \frac{e}{m_e} \langle F | \vec{A} \cdot \vec{p} | i⟩ ,
\]

where \( e \) and \( m_e \) are the charge and mass of a bare electron, \( \vec{A} \) is the vector potential of light and \( \vec{p} \) is the momentum operator defined as \( \vec{p} = (m_e/ih)[\vec{r},H] \). Here we assume \( \tau \) is the relaxation time for all bands.

Now we consider the light as

\[
E = \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} = \begin{pmatrix} \frac{i}{2} \sin(2\theta) \sin(\alpha) + (1 - i \cos(2\theta)) \cos(\phi) \cos(\alpha) \\ -i \sin(2\theta) \sin(\alpha) + (1 - i \cos(2\theta)) \cos(\phi) \cos(\alpha) \\ -\frac{i}{2} \cos(2\theta) \cos(\phi) \end{pmatrix},
\]

(34)

with \( E_0 \) as the magnitude of the electric field and \( \vec{A} = \vec{E}/i\omega \). Therefore under this polarized light \( |D|^2 \) is given as

\[
|D|^2 = \left( \frac{\omega e}{m_e} \right)^2 \left| \langle F | E_x p_x + E_y p_y + E_z p_z | i⟩ \right|^2 .
\]

(35)

We further rewrite \( E_aE^*_b \) (the subindices refer to Cartesian coordinates) as

\[
E_aE^*_b = [E_aE^*_b] + [E_aE^*_b] ,
\]

(36)

where \( \epsilon_{sab}[E_aE^*_b] = \frac{1}{2} (\vec{E} \times \vec{E}^*)_s \), with

\[
(\vec{E} \times \vec{E}^*)_x = -2i \cos(\alpha) \sin(2\theta) \sin(\phi)E_0^2 ;
\]

(37)

\[
(\vec{E} \times \vec{E}^*)_y = -2i \sin(\alpha) \sin(2\theta) \sin(\phi)E_0^2 ;
\]

(38)

\[
(\vec{E} \times \vec{E}^*)_z = -2i \sin(2\theta) \cos(\phi)E_0^2 .
\]

(39)

In the following, we drop \([E_aE^*_b]\) and \([E_a]^2\) terms which do not contribute to the circular photogalvanic effect, characterized by the \( \sin(2\theta) \) dependence. Therefore, we obtain

\[
|D|^2 = \frac{1}{2} \left( \frac{\omega e}{m_e} \right)^2 \left[ (\vec{E} \times \vec{E}^*)_x (\langle F | p_x | i⟩⟨i | p_y | F⟩ - ⟨F | p_y | i⟩⟨i | p_x | F⟩) 
- (\vec{E} \times \vec{E}^*)_y (⟨F | p_x | i⟩⟨i | p_z | F⟩ - ⟨F | p_z | i⟩⟨i | p_x | F⟩) 
+ (\vec{E} \times \vec{E}^*)_z (⟨F | p_y | i⟩⟨i | p_z | F⟩ - ⟨F | p_z | i⟩⟨i | p_y | F⟩) \right] .
\]

(40)

Here, the \( \sin(2\theta) \) factor that governs the currents changes sign when the helicity of light is inverted.
In a 2D crystal, we can use Peierls substitution, \( p_i = \frac{m_e}{\hbar} [z, \hat{H}] \) for \( i = x, y \). Because translational symmetry is broken along the \( z \) direction for a 2D crystal, we use \( p_z = \imath m_e / \hbar [z, \hat{H}] \) in \( |D|^2 \),

\[
|D|^2 = \left( \frac{\omega E_0 e^2}{\hbar} \right)^2 \left[ -\imath \cos(\phi) \sin(2\theta) \left( \langle F | \frac{\partial \hat{H}}{\partial k_x} | I \rangle \langle I | \frac{\partial \hat{H}}{\partial k_y} | F \rangle - \langle F | \frac{\partial \hat{H}}{\partial k_y} | I \rangle \langle I | \frac{\partial \hat{H}}{\partial k_x} | F \rangle \right) + \sin(\alpha) \sin(\phi) \sin(2\theta) \left( \langle F | \frac{\partial \hat{H}}{\partial k_x} | I \rangle \langle I | [z, \hat{H}] | F \rangle - \langle F | [z, \hat{H}] | I \rangle \langle I | \frac{\partial \hat{H}}{\partial k_x} | F \rangle \right) \right] \tag{41}
\]

The first term in \( |D|^2 \) can be related to Berry curvature (BC) for the electronic Bloch states of the \( n \)th band:

\[
\Omega^n_\alpha(\vec{k}) = i \hat{z} \cdot (\nabla_{\vec{k}} u^*_{nk}) \times (\nabla_{\vec{k}} u_{nk}) = -2 \sum_{n \neq n'} \frac{\text{Im} \left( \langle u_{nk} | \frac{\partial \hat{H}}{\partial k_x} | u_{n'k} \rangle \langle u_{n'k} | \frac{\partial \hat{H}}{\partial k_y} | u_{nk} \rangle \right)}{[E_n(\vec{k}) - E_{n'}(\vec{k})]^2} \tag{42}
\]

In a 2D crystal the Berry curvature has only a nonzero component, perpendicular to the \( xy \) plane (the Berry curvature behaves as a pseudoscalar). In a \( N \)-band system, the BC of the \( n \)th band comes from all the other \( N - 1 \) bands. Therefore the photocurrent Eq. (32) needs to sum over all possible initial and final states that satisfy the energy conservation \( \delta(\Delta E_{FI} - \omega) \). For a simple two-band approximation, \( F \) stands for the conduction band (CB) and \( I \) for the valence band (VB) with the definition of Berry curvature,

\[
\Omega^\alpha_F(\vec{k}) = -\Omega^\alpha_I(\vec{k}) = \frac{2 \text{Im} \left( \langle CB | \frac{\partial \hat{H}}{\partial k_x} | VB \rangle \langle VB | \frac{\partial \hat{H}}{\partial k_y} | CB \rangle \right)}{[E_{CB}(\vec{k}) - E_{VB}(\vec{k})]^2} \tag{43}
\]

This approximation allows us to simplify \( |D|^2 \) as

\[
|D|^2 = \left( \frac{\omega E_0 e^2}{\hbar} \right)^2 \left[ -\imath \cos(\phi) \sin(2\theta) \Omega^\alpha_F(\vec{k}) \Delta E_{FI} \right]^2 \\
+ \sin(\alpha) \sin(\phi) \sin(2\theta) \left( \langle F | \frac{\partial \hat{H}}{\partial k_x} | I \rangle \langle I | z | F \rangle - \langle F | z | I \rangle \langle I | \frac{\partial \hat{H}}{\partial k_x} | F \rangle \right) \Delta E_{FI} \tag{44}
\]

\[
- \cos(\alpha) \sin(\phi) \sin(2\theta) \left( \langle F | \frac{\partial \hat{H}}{\partial k_y} | I \rangle \langle I | z | F \rangle - \langle F | z | I \rangle \langle I | \frac{\partial \hat{H}}{\partial k_y} | F \rangle \right) \Delta E_{FI} 
\]

For multi-band cases, \( \Omega^\alpha_F(\vec{k}) \neq \Omega^\alpha_I(\vec{k}) \) and we could have additional corrections from other bands to the Berry curvature [4] of the forms
In pristine monolayer TMDCs, the conduction and valence band edges can be well-described by a two-band (without the inclusion of spins) or four-band (with the inclusion of spins), massive Dirac fermion model (see ref. 11 and, for a recent review, ref. 12). In this effective model (supported by first principle calculations), the Berry curvatures at the conduction and valence bands take opposite values, $\Omega_F^z(k) = -\Omega_I^z(k)$, where F stands for the conduction bands (CB) and I for the valence band (VB). The Berry curvature is dependent on spin through the spin-dependent band gap. In this description, the Fermi golden rule with two-band approximation allows us to connect the photocurrent to the Berry curvature with no additional corrections. TMDCs with lower symmetry are well described by a tilted Dirac model 13 and the Berry curvature is also found to take opposite values for conduction and valence band edges in this case. However, first principle calculations are needed for understanding the higher band corrections to the photocurrent.

One could also consider a possible role for intraband transitions, as discussed in ref. 14. However, these intraband contributions can only occur for $\hbar \omega < 2 \mu$, where $\hbar \omega$ is the the photon energy and $\mu$ is the chemical potential (choosing $\mu = 0$ at the bottom of the conduction band). In our measurements, the photon energy is around 1.6 eV, well above the chemical potential, in the order of 0.1-0.2 eV even when a gate voltage is applied. Thus, such effects are not expected to give a strong contribution here, but could still be observed in infrared to microwave regime.

In equation (44) the first term of $|D^2|$ is from Berry curvature $\Omega_F^z(k)$ and shows that this contribution to the CPGE is independent of $\alpha$, and maximal for normal incidence, $\phi = 0$. As discussed in the main text and in Supplementary Section 4, we do not find any contribution to CPC that satisfies this angular dependence. It is worth noting that, from the general definition of $j_{CPGE}$, equation (12), we find that a CPGE contribution changing as $\cos(\phi) \sin(2\theta)$ is associated with the matrix elements $\gamma_{xz}$ and $\gamma_{yz}$. The symmetry arguments discussed above confirm that these matrix elements can only be nonzero if the device symmetry is reduced to, at most, a single mirror plane. Therefore, the $D_{3h}$ symmetry of 1L-MoSe2 must be reduced (for example from device asymmetries or strain gradients) in order to allow for a Berry curvature-induced CPGE (BC-CPGE).
Supplementary Note 7: Spectral characterization of CPC amplitude for different voltages

Supplementary Figure 7 shows the CPC amplitude $C$ as a function of the illumination wavelength for different combinations of voltages, $V_{ds}$ and $V_g$. The resonant character of $C$ is clearly observed for all measurements, with the maximum signal occurring at 785 nm for $V_{ds} = 1$ V and at 790 nm for $V_{ds} = 0$ V (see also Fig. 3c in the main text).

For the data acquired at $V_{ds} = 1$ V, a weaker but nonzero CPC is also observed for off-resonance excitation with energies above the 1L-MoSe$_2$ absorption edge, which could be associated to the emergence of free-electron driven CPC. At negative gate voltages, this CPC contribution even presents a different sign from that of the main peak at $\sim 785$ nm. Although relevant, a comprehensive analysis of this off-resonance CPC contribution is beyond the scope of our present work.

**Supplementary Figure 7** Spectral dependence of $C$ at gate voltages, $V_g = 40$ V, -20 V and -40 V, as labelled, for $V_{ds} = 1$ V (a) and $V_{ds} = 0$ V (b). The spectral measurements corresponding to $V_g = 0$ can be found in the main text (Figure 3c). The dashed vertical lines are guides to the eye for 785 nm (green) and 790 nm (orange).
Supplementary Note 8: Brief note on valley exciton transitions in monolayer MoSe₂

The diagram shown in Supplementary Figure 8 summarizes the different excitonic transitions at the band edges of the K valley that can occur for monolayer MoSe₂ (similar transitions occur at the K’ valley, but all the spins have opposite orientation). For each pair of spin-orbit split subbands one gets an optically active neutral exciton ($A^0$ and $B^0$) which, at room temperature, is expected to give absorption peaks at 1.58 eV and 1.78 eV. Positively and negatively charged trion absorption can also occur ($A^{\pm/\pm}$ and $B^{\pm/\pm}$), at roughly 30 meV lower photon energies. Finally, electrons and holes from subbands with opposite spin can also combine to form the so-called dark excitons ($A_D^0$ and $B_D^0$). Dark exciton absorption is a priori spin-forbidden, but can become allowed for oblique illumination or even for normal incidence in the presence of a gate voltage. Further review on exciton physics in TMDCs can be found in references 17 and 18.

Supplementary Figure 8 Diagram illustrating the possible valley exciton transitions for monolayer MoSe₂.