

Nonlinear circular dichroism reveals the local Berry curvature

Received: 2 April 2026

Accepted: 4 September 2026

Published online: 13 October 2026

 Check for updates

Nele Tornow¹, Paul Herrmann¹, Clemens Schneider², Ferdinand Evers^{2,3},
Jan Wilhelm^{2,3}✉ & Giancarlo Soavi^{1,4}✉

The transfer of angular momentum between light and crystalline solids during nonlinear optical processes is typically interpreted in terms of angular momentum conservation and the rotational symmetry of the crystal lattice. However, these cornerstone principles rely on macroscopic symmetry arguments, providing little insight into the microscopic origin of angular momentum transfer. Here we show that changes in the spin angular momentum of light in high-harmonic generation processes, which we probe via nonlinear circular dichroism, are directly proportional to the local Berry curvature at a single optical resonance, therefore establishing a link between nonlinear light–matter interactions and the quantum geometry of electronic states. We demonstrate this relationship using second-harmonic generation in monolayer WSe₂, using circularly polarized light to break time-reversal symmetry and lift the degeneracy between the valleys. The resulting second-harmonic circular dichroism can be controlled through the intensity and helicity of the circularly polarized light, hence offering a method for the control and readout of the optically probed Berry curvature.

Inside a crystal, electrons accelerated by an electric field acquire a longitudinal velocity parallel to the field itself, with an amplitude that depends on the dispersion of the energy bands. In the presence of a magnetic field, the motion of electrons is further modified by the Lorentz force, resulting in a transverse (Hall) velocity perpendicular to the electric field. Remarkably, electrons can acquire a transverse velocity even without a magnetic field if they travel a portion of the reciprocal space where a non-zero Berry curvature is present. In this case, the velocity of an electron wave-packet reads $\mathbf{v} = \frac{1}{\hbar} \nabla_{\mathbf{k}} E_n(\mathbf{k}) + \dot{\mathbf{k}} \times \mathbf{\Omega}(\mathbf{k})$, where the first term describes the longitudinal velocity for a band with dispersion $E_n(\mathbf{k})$, $\dot{\mathbf{k}}$ is the time derivative of the crystal momentum (and it is parallel to the applied electric field) and $\mathbf{\Omega}(\mathbf{k})$ is the Berry curvature. The anomalous velocity defined by the Berry curvature is responsible for a circular motion of the electron wave-packet, and it is thus natural to ask whether there is a link between the Berry curvature and the angular momentum conservation laws in light–matter interactions, as the electric field of light can drive both the longitudinal and transverse velocities. Furthermore, the Berry curvature is intimately linked to

space inversion and time-reversal symmetries (TRSs), and thus to the valley physics¹: space inversion symmetry requires $\mathbf{\Omega}(\mathbf{k}) = \mathbf{\Omega}(-\mathbf{k})$, whereas TRS requires $\mathbf{\Omega}(\mathbf{k}) = -\mathbf{\Omega}(-\mathbf{k})$. Consequently, Berry curvature and the associated anomalous velocity play a central role in valleytronics and related transport and optical phenomena. More broadly, the Berry curvature constitutes the off-diagonal part of the quantum geometric tensor and, together with the quantum metric and their momentum-space derivatives (dipoles), is a fundamental property of electrons in crystals. These quantities underlie a wide range of optical and photocurrent phenomena, including circular dichroism (CD) in time-invariant crystals², valley-contrasted optical selection rules³ and the valley Hall effect⁴, the nonlinear Hall effect^{5–8}, and the bulk photovoltaic effect^{9,10}. The $\mathbf{k}$ -resolved Berry curvature is straightforward to compute from matrix elements of Bloch wavefunctions¹¹, but a direct experimental measurement of this fundamental quantity is extremely challenging. On the one hand, seminal theoretical and experimental studies have given a microscopic interpretation of the photoluminescence (PL) valley CD based on the valley-contrasting Berry curvature^{12–15},

¹Institute of Solid State Physics, Friedrich Schiller University Jena, Jena, Germany. ²Institute of Theoretical Physics and Regensburg Center for Ultrafast Nanoscopy, University of Regensburg, Regensburg, Germany. ³Halle-Berlin-Regensburg Cluster of Excellence CCE, Regensburg, Germany. ⁴Abbe Center of Photonics, Friedrich Schiller University Jena, Jena, Germany. ✉e-mail: jan.wilhelm@physik.uni-regensburg.de; giancarlo.soavi@uni-jena.de

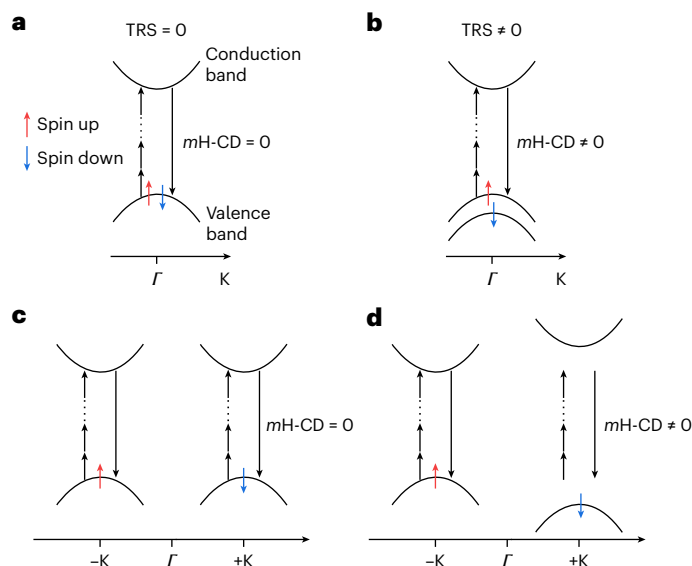


Fig. 1 | Examples of mH -CD. **a**, Spin-up (upward-pointing red arrow) and spin-down (downward-pointing blue arrow) energy bands are spin degenerate in Γ when both space inversion and TRSs are preserved. **b**, The breaking of TRS lifts the spin degeneracy, allowing a non-zero mH -CD. If the spin splitting is sufficiently large, the system can be modelled using equation (1). **c, d**, For crystals where space inversion symmetry is broken and TRS is preserved, energy degenerate levels at opposite momenta must have opposite spin (**c**). Breaking of TRS lifts the energy degeneracy between the valleys, allowing a non-zero mH -CD (**d**).

while on the other, recent works in the field of high-harmonic generation (HHG) have explored the interplay between Berry curvature and strong-field nonlinear optics^{16,17}, often using transition metal dichalcogenides (TMDs) as model systems. However, none of these works provided an analytical expression to quantitatively relate an experimentally accessible all-optical response to the Berry curvature, and, so far, only spin-resolved CD angle-resolved photoemission spectroscopy has been proposed as a direct measurement to this end^{18–21}, even though the range of validity and the resulting limitations of this approach are currently under discussion²². In this context, a quantitative, broadly accessible tabletop, all-optical probe of the Berry curvature with ultrafast time resolution could provide useful insights to our understanding of light–matter interactions and quantum geometry. Here we introduce such an approach via the nonlinear CD in harmonic generation. We show theoretically that for a single resonant crystal momentum the nonlinear CD constitutes a direct probe of the local Berry curvature, which further connects to the angular momentum conservation laws. We furthermore extend the model to the realistic case of multiple $\mathbf{k}$ -resonant optical transitions, and discuss nonlinear CD in relation to the local Berry curvature when TRS is broken. Finally, we test the predictions of our theory with time-resolved second-

harmonic generation (SHG) experiments in a prototypical WSe₂ monolayer TMD, where we break TRS by the coherent and valley-exclusive optical Stark and Bloch–Siegert effects^{23,24}. With this, we measure a value of the Berry curvature of $6.2 \pm 1.4 \text{ \AA}^2$, in good agreement with theoretical results from tight-binding²⁵ and density functional theory²⁶, and we thus establish second-harmonic CD (SH-CD) as an all-optical, ultrafast and non-invasive probe of the local Berry curvature.

SH-CD and Berry curvature

We work in an independent-particle two-band picture considering a single vertical transition at a well-defined crystal momentum $\mathbf{k}_{\text{res}}$ and we treat $\varepsilon_{cv}(\mathbf{k}_{\text{res}})$ as an effective resonance energy. At resonance, energy conservation reads $\varepsilon_{cv}(\mathbf{k}_{\text{res}}) = m\hbar\omega$, and the m th harmonic CD

(mH -CD) directly probes the local Berry curvature $\Omega(\mathbf{k}_{\text{res}})$ (Supplementary Section 1)

$$mH\text{-CD} = \frac{|P^+(m\omega)|^2 - |P^-(m\omega)|^2}{|P^+(m\omega)|^2 + |P^-(m\omega)|^2} = \frac{\Omega(\mathbf{k}_{\text{res}})}{\|\mathbf{d}_{vc}(\mathbf{k}_{\text{res}})\|^2}, \quad (1)$$

where $P^\pm(m\omega)$ is the circular harmonic polarization and $\mathbf{d}_{vc}(\mathbf{k}_{\text{res}})$ is the interband dipole matrix element. This equation is general and does not impose any symmetry-related restrictions, provided that the relevant energy levels are sufficiently well separated to prevent overlapping resonances involving multiple $\mathbf{k}$ -points. In this scenario, light–matter interactions can probe individual energy bands and their spin $E_\uparrow$ and $E_\downarrow$. We furthermore highlight that $E_\uparrow(+\mathbf{k}) = E_\uparrow(-\mathbf{k})$ when space inversion symmetry is preserved, and $E_\uparrow(+\mathbf{k}) = E_\downarrow(-\mathbf{k})$ when TRS is preserved. Thus, the requirements to fulfil equation (1) are only the existence of the relevant harmonics (for example, even harmonics are forbidden in the electric dipole approximation when space inversion symmetry is preserved) and the presence of a single optically accessible resonance in $\mathbf{k}$ -space, which can be treated as an effective two-level system. This can be readily achieved by TRS breaking either in a single optical resonance at Γ (Fig. 1a,b) or in valleys at opposite momenta in reciprocal space (Fig. 1c,d). It is worth noting that this equation is also not restricted to direct-gap semiconductors, but it applies generally whenever an optically accessible vertical transition gives rise to a resonant HHG response²⁷. This formalism extends the finding of linear optics, $m=1$ (refs. 12,13,28), and it directly links mH -CD to the interband polarization rotation and to the angular momentum conservation law in light–matter interactions.

Let us now consider a linearly polarized probe beam resonant with ε_{cv} impinging on the sample and generating high harmonics with a different efficiency for left and right helicities, namely a non-zero mH -CD. In this case, an initial zero spin angular momentum of light (linear polarization) converts into a non-zero spin angular momentum in HHG. This example readily highlights the direct link between local Berry curvature for a single optical resonance and angular momentum conservation in nonlinear optics. A more realistic situation, involving two energetically degenerate resonances at opposite momenta $\pm\mathbf{K}$, can be experimentally realized in monolayer TMDs, for which we derive a detailed analytical expression of the SH-CD. An off-resonant circularly polarized pump beam lifts the energy degeneracy in the $\pm\mathbf{K}$ valleys and, subsequently, a probe beam with photon energy close to half of the optical band gap generates a resonant SH signal. As discussed, the change in spin angular momentum during the nonlinear process is captured by the SH-CD, defined as

$$\text{SH-CD} = \frac{|\chi_{+\beta\beta}^{(2)}|^2 - |\chi_{-\beta\beta}^{(2)}|^2}{|\chi_{+\beta\beta}^{(2)}|^2 + |\chi_{-\beta\beta}^{(2)}|^2}, \quad (2)$$

where $\chi_{\sigma\beta\beta}^{(2)}$ is the second order nonlinear susceptibility of a $\sigma = \pm$ circularly polarized SH signal, given a linearly polarized probe beam along the generic Cartesian axis β . If the pump beam is switched off, the energy degeneracy in the $\pm\mathbf{K}$ valleys is preserved (Fig. 2a,b), the SH-CD vanishes and a linear input probe beam generates a linear SH output. If the pump beam is so intense that the two valleys become highly non-degenerate (Fig. 2c,d) then the SH-CD is again described by equation (1), as for a single optical resonance. For an intermediate situation with a small (~ 10 meV for typical experimental conditions^{23,24}) energy difference between the valleys (Fig. 2e,f), the SH-CD can be calculated analytically by including the valley-dependent optical Stark and Bloch–Siegert shifts into the two-band Hamiltonian from refs. 23,29, leading to (see Supplementary Section 2 for details)

$$\begin{aligned} \text{SH-CD} &= \frac{\sum_{\mathbf{k}=\pm\mathbf{K}} w(\mathbf{k}, \varepsilon_0, \omega) \Omega(\mathbf{k}, \varepsilon_0)}{\sum_{\mathbf{k}=\pm\mathbf{K}} w(\mathbf{k}, \varepsilon_0, \omega) \|\mathbf{d}_{vc}(\mathbf{k}, \varepsilon_0)\|^2} \\ &= 2|\Omega(\pm\mathbf{K})| \sigma \varepsilon_0^2 g(v) f(\omega) + \mathcal{O}(\varepsilon_0^4), \end{aligned} \quad (3)$$

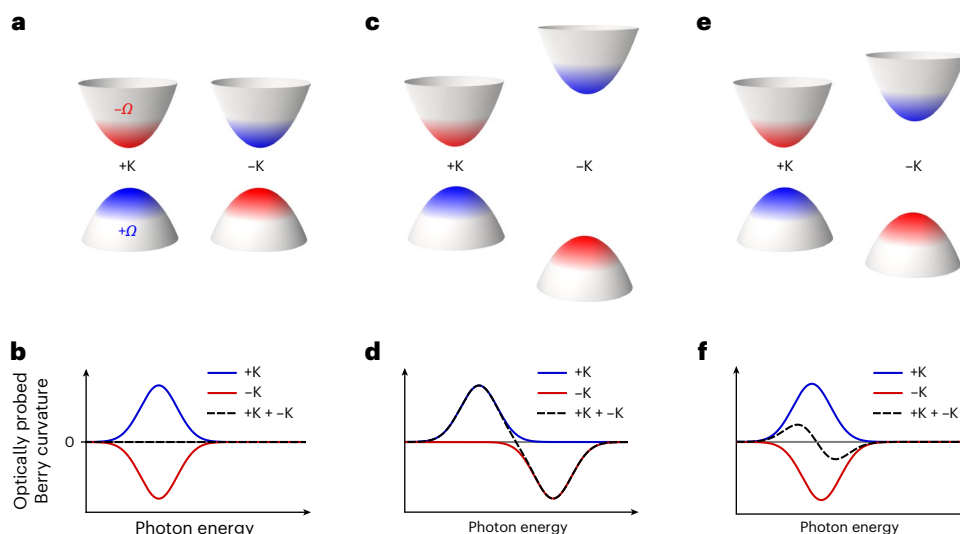


Fig. 2 | Valleys and optically probed Berry curvature in monolayer TMDs.

a,c,e. The equilibrium (**a**) and TRS broken (**c** and **e**) schematic band structure and Berry curvature $\pm\Omega$ of a monolayer TMD. Red and blue areas indicate Berry curvature with opposite signs. **c,e.** A large shift in the bands yields a single resonance (**c**) while a smaller shift allows light to probe both valleys (**e**).

b,d,f. Sketches of the optically probed valence-band Berry curvature in each valley (red/blue curves) and their sum (dashed black line) at equilibrium and for broken TRS. The sum of the optically probed Berry curvature at $\pm K$ defines the strength of the SH-CD, as captured by equation (3).

where $w(\mathbf{k}, \varepsilon_0, \omega)$ is a positive weight that contains the valley-resolved resonance denominators, $\Omega(\mathbf{k}, \varepsilon_0)$ is the Berry curvature dressed by the pump beam field of amplitude ε_0 , and $\mathbf{d}_{vc}(\mathbf{k}, \varepsilon_0)$ is the corresponding interband dipole matrix element. We identify the product $w(\mathbf{k}, \varepsilon_0, \omega) \Omega(\mathbf{k}, \varepsilon_0)$ as the optically probed Berry curvature, that we measure in our experiments (Fig. 2b,d,f).

Equation (3) directly shows the dependence of the SH-CD on the absolute value of the Berry curvature of each unperturbed valley $\Omega(\pm K)$, on the intensity of the pump field ($\sim \varepsilon_0^2$), which is a direct consequence of TRS breaking via all-optical coherent effects^{23,27}, and the sign reversal of the SH-CD upon changing the helicity $\sigma = \pm 1$ of the pump beam. The remaining proportionality factors in equation (3) are $g(v) = v/(\Delta^2(1 - v^2))$, where $v = \omega_{\text{pump}}/\Delta$ is the normalized pump beam frequency and Δ is the equilibrium direct gap, and $f(\omega) = 2 + \text{Re} \eta(\hbar\omega)$, which describes the detuning of the probe beam frequency ω from the equilibrium SH resonance at $\hbar\omega = \Delta/2$ via the complex resonance factor $\eta(\hbar\omega) = \Delta/(\Delta - 2\hbar\omega - i\hbar/T_2)$, where T_2 is the dephasing time of the optical resonance.

Considering a probe beam propagating along the z crystallographic direction and with polarization in the x crystallographic direction, the SH-CD can be further expressed as

$$\text{SH-CD} = \frac{2\text{Im}[\chi_{xxx}^{(2)}(\chi_{yxx}^{(2)})^*]}{|\chi_{xxx}^{(2)}|^2 + |\chi_{yxx}^{(2)}|^2}, \quad (4)$$

indicating that SH-CD can, in principle, only manifest in the point groups 1, 3 and $\bar{6}$ if TRS is preserved, where circular SHG is allowed by momentum conservation and both $\chi_{xxx}^{(2)}$ and $\chi_{yxx}^{(2)}$ are non-zero³⁰. However, it is interesting to note that a further requirement for the SH-CD to be non-zero is that $\chi_{xxx}^{(2)}$ and $\chi_{yxx}^{(2)}$ are not in phase, a property that cannot be deduced from a simple macroscopic description of the nonlinear tensors. When TRS is broken, and considering the admissible spin directions^{27,31}, SH-CD can be non-zero for all non-centrosymmetric magnetic groups with 1- and 3-fold rotational symmetry, that is 1, 2', m' , 3, $3m'$, $32'$, $\bar{6}$ and $\bar{6}m'2'$, where the latter includes the case of monolayer TMDs with broken TRS. For all other magnetic point groups, circular SHG is forbidden either by parity (centrosymmetric crystals), or by angular momentum conservation.

Equation (4), combined with equation (3), indicates that an asymmetry between energy levels ($E_v(+K) \neq E_v(-K)$ and $E_v(+K) \neq E_v(-K)$) and Berry curvature ($\Omega(+K) \neq \Omega(-K)$ and $\Omega(+K) \neq -\Omega(-K)$) at opposite momenta in reciprocal space induces a phase mismatch between the emerging $\chi_{xxx}^{(2)}$ and $\chi_{yxx}^{(2)}$ components of the second order susceptibility. This provides physical insights that link macroscopic symmetries to microscopic features of the electronic band structure and quantum geometry.

Ultrafast all-optical control of nonlinear CD

We demonstrate ultrafast all-optical control of the SH-CD in a two-colour pump-probe experiment in a WSe₂ monolayer. Exemplary helicity resolved emission spectra are shown in Fig. 3 for zero and long (that is, much longer than the pulse duration) delays between a left circularly polarized (LCP) pump beam and a linearly polarized probe beam, responsible for SH generation. The pump beam/probe beam wavelengths are fixed at 1,800 nm/1,460 nm (see Methods for details). In Fig. 3, the black/red curves represent spectra with co-/cross-polarization with respect to the circular pump beam, while the blue curves show their difference. In all cross-polarized spectra, regardless of the pump beam to probe beam time delay, we observe the SH of the pump beam at 900 nm, as expected from spin angular momentum conservation³². This signal is absent in the co-polarized spectra. At zero delay (Fig. 3b), we further observe a sum frequency generation (SFG) signal between the pump beam and probe beam, which is again cross-polarized with respect to the pump beam. In the region between approximately 730 nm and 750 nm, we observe the most relevant signal for our study of SH-CD, namely the SH of the probe beam. This overlaps spectrally with the two-photon PL (TP-PL), mainly originating from the probe beam. When taking the difference between LCP and right circularly polarized (RCP) detection (Fig. 3, blue curves), the TP-PL always cancels out owing to its incoherent nature at room temperature³³. By contrast, the probe beam second harmonic (probe SH) signal appears clearly in the RCP-LCP spectrum at zero delay, underpinning the SH-CD. The signal is drastically reduced at large delays, where the small residual is most likely due to an initial and uncompensated helicity of the probe beam. With this experimental scheme, we can further modulate the SH-CD by tuning the pump beam to probe beam time delay and the pump beam

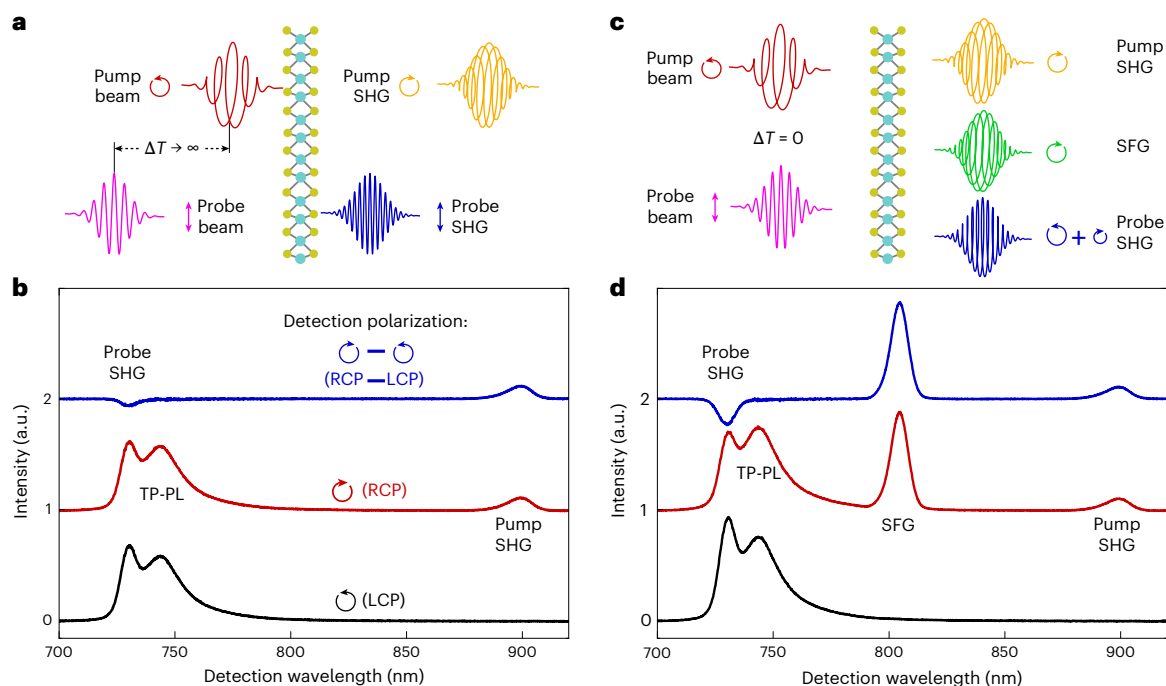


Fig. 3 | Helicity and time-resolved second-harmonic spectra. a, A large negative delay ($\Delta T \rightarrow \infty$) between the LCP pump beam (red pulse) and a linearly polarized probe beam (pink pulse) leads to the emission of RCP SHG of the pump pulse and linearly polarized SHG of the probe beam (blue pulse). **b**, LCP (black curve) and RCP (red curve) resolved emission spectra of the experiment in **a** both show similar SHG intensity of the probe beam and additional TP-PL. Both are vanishing in the difference spectrum (blue curve). Pump beam SHG appears only in the RCP spectrum and therefore in the difference spectrum as

well. **c**, LCP pump beam (red) and linearly polarized probe beam (pink) now at zero time delay ($\Delta T = 0$) cause RCP pump beam SHG (yellow) and SFG (green), as well as elliptically polarized SHG of the probe beam (blue). **d**, LCP (black) and RCP (red) spectra at zero time delay feature a difference in intensity of probe beam SHG, also leading to a peak in the difference spectrum (blue), indicating a non-zero SH-CD. RCP SFG and SHG of the pump beam appear only in the RCP and difference spectrum.

intensity (Fig. 4). On the basis of equation (3), this provides a possible route towards the realization of ultrafast all-optical devices combining the valley degree of freedom with the engineering and readout of the local Berry curvature. Figure 4a shows the ultrafast dynamical change of the SH-CD, which occurs on a time-scale defined only by the pump beam and probe beam pulse duration (≤ 300 fs). The time-dependent response can be fitted by a Gaussian peak with a full-width at half maximum of 313 ± 40 fs, limited by the time profile of the pulses used in our experiments. This ultrafast response time is inherent to the approach used for TRS breaking with below-gap off-resonant excitation. The sign change of the SH-CD by reversing the pump beam helicity corresponds to a change in the energy offset between the $\pm K$ valleys, as predicted also by the linear dependence with σ in equation (3). Furthermore, the SH-CD is independent of time delay for linear polarization of the pump beam (Fig. 4a, red triangles), namely when TRS is preserved. A further experimental tuning knob is the pump beam intensity: coherent off-resonant band gap modulation produces an asymmetry in the energy band gap of the $\pm K$ valleys that is linearly proportional to the light intensity, leading to the ε_0^2 dependence in equation (3) and in agreement with the experimental results in Fig. 4b. Finally, the experimental SH-CD as a function of the probe beam wavelength exhibits a sign reversal close to half the equilibrium band gap (Fig. 4c), although the circular polarization of the pump beam is kept fixed. This behaviour is due to tuning of the SH wavelength across the light-dressed $\pm K$ valleys, resulting in a modulation of the corresponding resonant contributions. Notably, this represents the TRS-broken counterpart of the effect reported in ref. 34 for linearly polarized excitation. Within our analytical framework, this mechanism is captured by the frequency-dependent function $f(\omega)$ in equation (3). The best fit of equation (3) to our experimental data (Fig. 4c, dashed grey line) provides a direct

estimate of the dephasing time $T_2 = 42 \pm 11$ fs and of the $\pm K$ resonant Berry curvature $|\Omega(\pm K)| = 6.2 \pm 1.4 \text{ \AA}^2$, in good agreement with reported values from tight-binding²⁵ and density functional theory²⁶ simulations. We highlight that T_2 and $|\Omega(\pm K)|$ are the only free parameters in our fit, as all other variables in equation (3) are experimentally measured and thus fixed (Supplementary Section 4).

Conclusions

We have shown that nonlinear CD is a powerful ultrafast and all-optical method to reveal the local Berry curvature at optical resonances. Conservation of angular momentum always imposes that the momentum transfer from the electromagnetic field to the crystal lattice is quantized in multiple integers of $r\hbar$, where r is the discrete rotational symmetry of the crystal. In addition to this fundamental conservation law, we demonstrate that the optically probed Berry curvature underlies the efficiency at which photons with opposite helicity are generated, resulting in a net nonlinear CD. We derived an analytical expression of this general model for a prototype atomically thin semiconductor with threefold rotational symmetry, focusing in particular on its SH-CD upon breaking of TRS with light-dressed states. This provides physical insights into the microscopic origin of the elements of the nonlinear optical susceptibility, and their link to the local quantum geometry. Finally, our analytical model qualitatively and quantitatively describes experimental results of the dynamical SH-CD in a monolayer WSe₂ sample, performed in a pump-probe configuration where the time delay, pump beam intensity and helicity, and probe beam wavelength are used as tuning knobs. This work offers an approach for the nonlinear all-optical readout of the local quantum geometry, and it paves the way to ultrafast all-optical devices combining the spin and valley degrees of freedom to the local Berry curvature, thus advancing the field of ultrafast all-optical valleytronics^{1,35–38}.

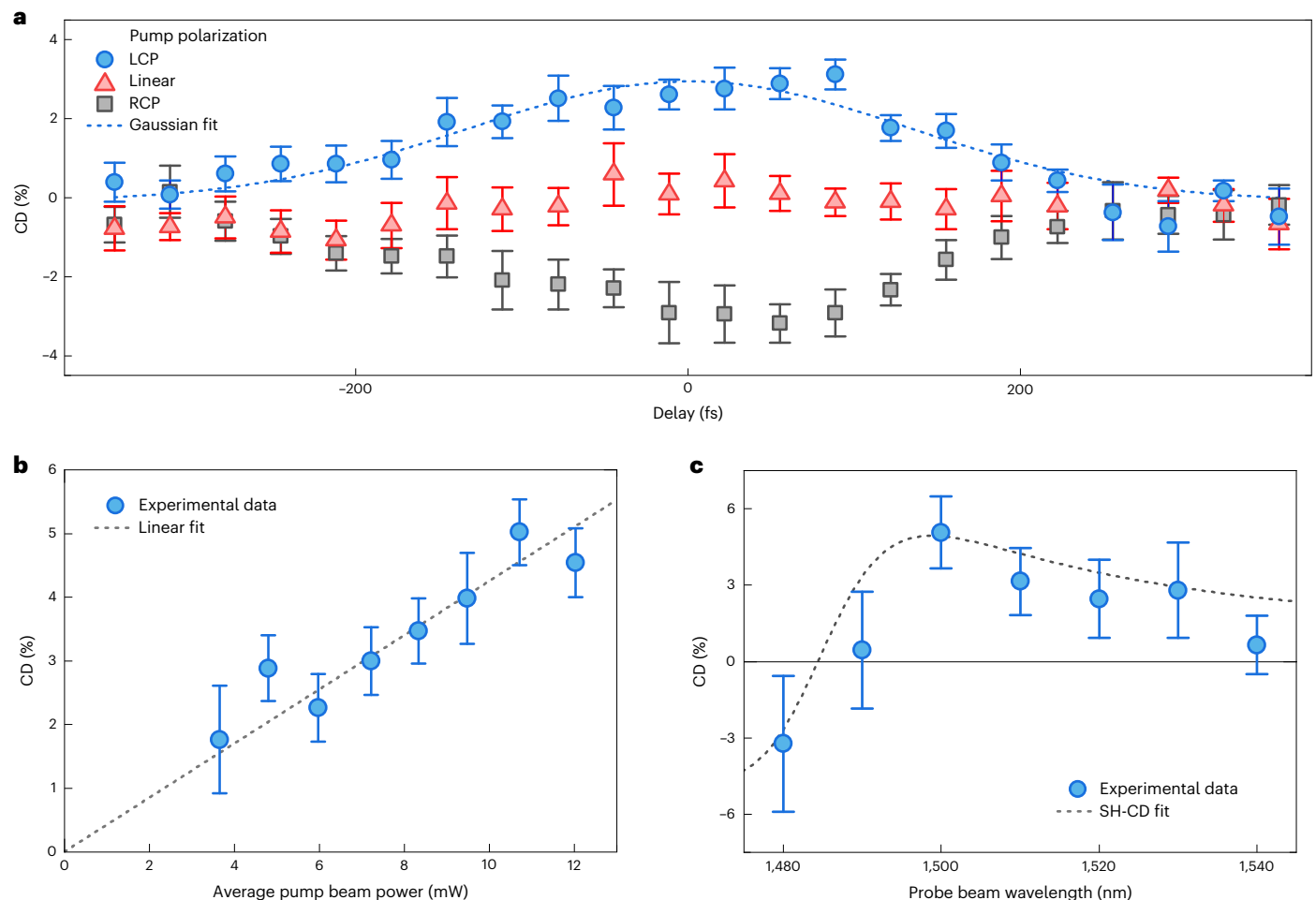


Fig. 4 | Experimental tuning of the SH-CD. a, Delay scans of the SH-CD for different pump beam polarizations (circular and linear). Pump beam and probe beam wavelengths are 1,800 nm and 1,500 nm, respectively. The SH-CD is independent of time delay when the pump beam is linearly polarized (red triangles), while it changes with time delay and flips sign when the helicity of the circularly polarized pump beam is reversed. The dashed line represents a Gaussian fit with a full-width at half maximum of 313 ± 40 fs. Datapoints are shown as averages with their standard deviation. **b,** The pump beam power dependence of the SH-CD at zero time delay and for the LCP pump beam. The probe beam wavelength is fixed at 1,500 nm. The intercept of the linear fit

(dashed grey line) is set to zero. SH-CD values are the amplitudes of the Gaussian fits of time delay measurements as in **a**. The error bars are the fitted errors of the amplitude of the Gaussian fits. **c,** Probe beam wavelength dependence of the SH-CD. Equation (3) is used to fit the averaged experimental data (blue circles) including standard deviation using the experimental parameters pump beam electric field strength $\mathcal{E}_0 = 0.2 \text{ V nm}^{-1}$, equilibrium band gap $\Delta = 1.662 \text{ eV} + 8.2 \text{ meV}$ shift from optical dressing of the probe beam (Supplementary Section 3), and normalized pump beam frequency $\nu = 0.415$, leading to dephasing time $T_2 = 42 \pm 11 \text{ fs}$ and $\pm K$ resonant Berry curvature $|\Omega(\pm K)| = 6.2 \pm 1.4 \text{ \AA}^2$.

Online content

Any methods, additional references, Nature Portfolio reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at <https://doi.org/10.1038/s41566-026-02025-8>.

References

- Seyler, K. L. et al. Valleytronics in 2D materials roadmap. Preprint at <https://doi.org/10.48550/arXiv.2603.01427> (2026).
- Soavi, G. & Wilhelm, J. The role of Berry curvature derivatives in the optical activity of time-invariant crystals. Preprint at <https://doi.org/10.48550/arXiv.2501.03684> (2025).
- Li, Y. & Liu, C.-C. Quantum-metric-based optical selection rules. *Phys. Rev. Lett.* **136**, 046901 (2026).
- Mak, K. F., McGill, K. L., Park, J. & McEuen, P. L. Valleytronics. The valley Hall effect in MoS_2 transistors. *Science* **344**, 1489–1492 (2014).
- Sodemann, I. & Fu, L. Quantum nonlinear Hall effect induced by Berry curvature dipole in time-reversal invariant materials. *Phys. Rev. Lett.* **115**, 216806 (2015).
- Xu, S.-Y. et al. Electrically switchable Berry curvature dipole in the monolayer topological insulator WTe_2 . *Nat. Phys.* **14**, 900–906 (2018).
- Ma, Q. et al. Observation of the nonlinear Hall effect under time-reversal-symmetric conditions. *Nature* **565**, 337–342 (2019).
- Wang, H. & Qian, X. Ferroelectric nonlinear anomalous Hall effect in few-layer WTe_2 . *npj Comput. Mater.* **5**, 119 (2019).
- Ma, Q., Krishna Kumar, R., Xu, S.-Y., Koppens, F. H. L. & Song, J. C. W. Photocurrent as a multiphysics diagnostic of quantum materials. *Nat. Rev. Phys.* **5**, 170–184 (2023).
- Morimoto, T. & Nagaosa, N. Topological nature of nonlinear optical effects in solids. *Sci. Adv.* **2**, e1501524 (2016).
- Xiao, D., Chang, M.-C. & Niu, Q. Berry phase effects on electronic properties. *Rev. Mod. Phys.* **82**, 1959–2007 (2010).
- Yao, W., Xiao, D. & Niu, Q. Valley-dependent optoelectronics from inversion symmetry breaking. *Phys. Rev. B* **77**, 235406 (2008).
- Cao, T. et al. Valley-selective circular dichroism of monolayer molybdenum disulphide. *Nat. Commun.* **3**, 887 (2012).
- Wu, S. et al. Electrical tuning of valley magnetic moment through symmetry control in bilayer MoS_2 . *Nat. Phys.* **9**, 149–153 (2013).

15. Kim, S.-W., Kim, H.-J., Cheon, S. & Kim, T.-H. Circular dichroism of emergent chiral stacking orders in quasi-one-dimensional charge density waves. *Phys. Rev. Lett.* **128**, 046401 (2022).
16. Bae, G., Kim, Y. & Lee, J. D. Revealing Berry curvature of the unoccupied band in high harmonic generation. *Phys. Rev. B* **106**, 205422 (2022).
17. Uzan-Narovlansky, A. J. et al. Observation of interband Berry phase in laser-driven crystals. *Nature* **626**, 66–71 (2024).
18. Schüller, M. et al. Local Berry curvature signatures in dichroic angle-resolved photoelectron spectroscopy from two-dimensional materials. *Sci. Adv.* **6**, eaay2730 (2020).
19. Beaulieu, S. et al. Berry curvature signatures in chiroptical excitonic transitions. *Sci. Adv.* **10**, eadk3897 (2024).
20. Kim, S. et al. Direct measurement of the quantum metric tensor in solids. *Science* **388**, 1050–1054 (2025).
21. Kang, M. et al. Measurements of the quantum geometric tensor in solids. *Nat. Phys.* **21**, 110–117 (2025).
22. Sidilkover, I. et al. Reexamining circular dichroism in photoemission from a topological insulator. *Phys. Rev. Res.* **7**, 033027 (2025).
23. Herrmann, P. et al. Nonlinear valley selection rules and all-optical probe of broken time-reversal symmetry in monolayer WSe₂. *Nat. Photon.* **19**, 300–306 (2025).
24. Sie, E. J. et al. Large, valley-exclusive Bloch–Siegert shift in monolayer WS₂. *Science* **355**, 1066–1069 (2017).
25. Cho, S. et al. Experimental observation of hidden Berry curvature in inversion-symmetric bulk 2H-WSe₂. *Phys. Rev. Lett.* **121**, 186401 (2018).
26. Xiao, D., Liu, G.-B., Feng, W., Xu, X. & Yao, W. Coupled spin and valley physics in monolayers of MoS₂ and other group-VI dichalcogenides. *Phys. Rev. Lett.* **108**, 196802 (2012).
27. Friedrich, F. et al. Measurement of optically induced broken time-reversal symmetry in atomically thin crystals. *Nat. Photon.* **20**, 186–193 (2026).
28. Sekh, S. & Mandal, I. Circular dichroism as a probe for topology in three-dimensional semimetals. *Phys. Rev. B* **105**, 235403 (2022).
29. Taghizadeh, A. & Pedersen, T. G. Nonlinear optical selection rules of excitons in monolayer transition metal dichalcogenides. *Phys. Rev. B* **99**, 235433 (2019).
30. Boyd, R. W. *Nonlinear Optics* 4th edn (Academic Press, 2020).
31. Rodríguez-Carvajal, J. & Bourée, F. Symmetry and magnetic structures. *EPJ Web Conf.* **22**, 00010 (2012).
32. Bloembergen, N. Conservation laws in nonlinear optics. *J. Opt. Soc. Am.* **70**, 1429 (1980).
33. Herrmann, P. et al. Nonlinear all-optical coherent generation and read-out of valleys in atomically thin semiconductors. *Small* **19**, e2301126 (2023).
34. Klimmer, S. et al. Probing ultrafast coherent bandgap modulation in monolayer WSe₂ by nonlinear optics. *Adv. Opt. Mater.* **14**, e03236 (2026).
35. Gindl, A., Čmel, M., Trojánek, F., Malý, P. & Kozák, M. Ultrafast room-temperature valley manipulation in silicon and diamond. *Nat. Phys.* **21**, 947–952 (2025).
36. Gucci, F. et al. Encoding and manipulating ultrafast coherent valleytronic information with lightwaves. *Nat. Photon.* **20**, 266–272 (2026).
37. Tyulnev, I. et al. Valleytronics in bulk MoS₂ with a topologic optical field. *Nature* **628**, 746–751 (2024).
38. Mitra, S. et al. Light-wave-controlled Haldane model in monolayer hexagonal boron nitride. *Nature* **628**, 752–757 (2024).

Publisher's note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit <http://creativecommons.org/licenses/by/4.0/>.

© The Author(s) 2026

Methods

SH-CD measurements

For the two-colour SH-CD experiments, the pump beam and probe beam are taken from two synchronized optical parametric oscillators (Levante IR fs, A.P.E) pumped by an ytterbium-doped laser with a central wavelength at 1,030 nm, a repetition rate of 76 MHz and a pulse duration of ~100 fs (FLINT FL2-12, LIGHT CONVERSION). The time delay between pump beam and probe beam is tuned by a motorized delay stage (M-414.2PD, PI with controller C-863.12, PI). The probe beam polarization and power are controlled by a combination of a linear (WP25M-UB, Thorlabs) and a Glan–Thompson polarizer (GTH10M). The probe beam electric field strength was set to 0.16 V nm^{-1} . The power of the pump beam is controlled by the combination of two linear polarizers (WP25M-UB, Thorlabs). The polarization of the pump beam is tuned by combining a half-wave plate and quarter-wave plate (RSU1.2.15 and 1.4.15, B-Halle) in two motorized rotational mounts (PRM1/MZ8, Thorlabs). The pump beam and probe beam are subsequently combined at a Polkadot beamsplitter (BPD5254-G01, Thorlabs) and focused onto the sample with a $40\times$ reflective objective (LMM-40X-UVV, Thorlabs). After the sample, we use a lens (C330TMD, Thorlabs) and different sets of spectral filters to isolate the SH signal from the residual transmitted pump beam and probe beam. To measure the ellipticity and CD of the SH beam, we use a quarter-wave plate (63-935, Edmund optics) at $\pm 45^\circ$ with respect to a fixed linear polarizer (WP25M-UB, Thorlabs), which is aligned parallel to the SH of the probe beam. The spectra in Fig. 3 are taken with a monochromator (iHR320, HORIBA) combined with a nitrogen cooled silicon detector (Symphony II, HORIBA), and the probe beam is blocked by a shortpass filter (FESH0950, Thorlabs). For the measurements in Fig. 4a,b, we detect the SH signal with a silicon avalanche photodiode (APD440A, Thorlabs) and lock-in amplification (MFLI, Zurich Instruments), which filters the signal coming from the probe beam modulated at 967 Hz via an optical chopper (MC2000B, Thorlabs). Bandpass filters are chosen for each specific SH wavelength (FBHxxx-10, Thorlabs).

Sample preparation and characterization

The monolayer WSe_2 was mechanically exfoliated from a bulk crystal (HQ graphene) onto polydimethylsiloxane and transferred onto a fused silica substrate using a commercial transfer stage (HQ graphene). The monolayer nature of the sample was determined by PL measurements and a cross-polarized SH polarization pattern with linearly polarized probe beam was performed to rule out the presence of strain in the sample (Supplementary Section 5).

Error estimation for Berry curvature and dephasing time

For the estimation of $|\Omega(\pm K)|$ and T_2 , we used the experimental values, including the error bars in Fig. 4c, and fitted equation (3) to it. The deviation given for the values are the standard deviations of the average results of 5,000 fits performed, where for each data point a value following a Gaussian distribution around the mean value within the standard deviation is chosen for each fit performed.

Data availability

The data that support the plots within this paper and other findings of this study are available from the corresponding author on request. Source data are provided with this paper.

Acknowledgements

We thank M. Berry, A. Seith and S. Shanbhag for helpful discussions.

We thank S. Klimmer for valuable assistance with figure design and visualization.

Author contributions

G.S. and J.W. conceived the work. N.T. and P.H. prepared the sample, performed the measurements and analysed the data. N.T., P.H. and G.S. interpreted the experimental results. C.S., F.E. and J.W. developed the analytical model. The paper was written with contributions from all co-authors.

Funding

G.S. acknowledges funding from the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) via the SFB 1375 NOA ‘Nonlinear Optics down to Atomic scales’ (project number 398816777), IRTG 2675 ‘META-ACTIVE’ (project numbers 437527638 and 448835038) and WHAT-A-TWIST (project number 547611111). J.W. acknowledges the DFG for funding via the Emmy Noether Programme (project number 503985532), CRC 1277 (project number 314695032, subproject A03) and RTG 2905 (project number 502572516). F.E. acknowledges that this work was supported by the DFG under EV30/12-1, EV30/14-1, EV30/16-1, further CRC 1277 (project-ID: 314695032, subproject A03) and RTG 2905 (project number 502572516). J.W. and F.E. acknowledge the DFG through the German Excellence Strategy (EXC3112/1 - 533767171) (Center for Chiral Electronics). Open access funding provided by Universität Regensburg.

Competing interests

The authors declare no competing interests.

Additional information

Supplementary information The online version contains supplementary material available at <https://doi.org/10.1038/s41566-026-02025-8>.

Correspondence and requests for materials should be addressed to Jan Wilhelm or Giancarlo Soavi.

Peer review information *Nature Photonics* thanks the anonymous reviewer(s) for their contribution to the peer review of this work. Peer reviewer reports are available.

Reprints and permissions information is available at www.nature.com/reprints.