Giant nonreciprocal second-harmonic generation from antiferromagnetic bilayer CrI\(_3\)

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Layered antiferromagnetism is the spatial arrangement of ferromagnetic layers with antiferromagnetic interlayer coupling. The van der Waals magnet chromium triiodide (CrI\(_3\)) has been shown to be a layered antiferromagnetic insulator in its few-layer form\(^6\), opening up opportunities for various functionalities\(^5\) in electronic and optical devices. Here we report an emergent nonreciprocal second-order nonlinear optical effect in bilayer CrI\(_3\). The observed second-harmonic generation (SHG; a nonlinear optical process that converts two photons of the same frequency into one photon of twice the fundamental frequency) is several orders of magnitude larger than known magnetization-induced SHG\(^8–11\) and comparable to the SHG of the best (in terms of nonlinear susceptibility) two-dimensional nonlinear optical materials studied so far\(^12,13\) (for example, molybdenum disulfide). We show that although the parent lattice of bilayer CrI\(_3\) is centrosymmetric, and thus does not contribute to the SHG signal, the observed giant nonreciprocal SHG originates only from the layered antiferromagnetic order, which breaks both the spatial-inversion symmetry and the time-reversal symmetry. Furthermore, polarization-resolved measurements reveal underlying C\(_{2h}\) crystallographic symmetry—and thus monoclinic stacking order—in bilayer CrI\(_3\), providing key structural information for the microscopic origin of layered antiferromagnetism\(^8–11\). Our results indicate that SHG is a highly sensitive probe of subtle magnetic orders and open up possibilities for the use of two-dimensional magnets in nonlinear and nonreciprocal optical devices.

SHG is not only of technological importance for nonlinear optical devices, but also a powerful tool for the investigation of symmetry-related physical phenomena that are otherwise challenging to probe. The power of this technique lies in its sensitivity to inversion–symmetry breaking, which is the prerequisite for non-vanishing SHG under the electric dipole approximation. For a system without lattice inversion symmetry, SHG is thus electric-dipole-allowed and is known to be a time-invariant, or i-type, process. In the presence of lattice inversion symmetry, SHG can also be allowed if, for instance, there is an underlying magnetic structure that breaks both spatial-inversion and time-reversal symmetries\(^8–11\). This electric-dipole-allowed SHG becomes time-noninvariant or nonreciprocal, denoted as c-type. Compared to i-type SHG, c-type SHG is less common and often weaker, which has been used to probe antiferromagnetic order in bulk crystals such as Cr\(_2\)O\(_3\)\(^{9–11,19}\) and surface ferromagnetism in transition-metal thin films\(^8,20,21\).

The recent discovery of two-dimensional (2D) van der Waals magnets\(^{12–28}\) may provide a new platform for exploring second-order nonlinear optical effects. Among these magnets, bilayer CrI\(_3\) is particularly interesting because of the interplay between its crystal structure and magnetic order. As shown in Fig. 1a, monolayer CrI\(_3\) has a centrosymmetric lattice structure with three-fold rotational symmetry\(^4\). When two monolayer sheets are stacked along the same orientation (Fig. 1b), the bilayer CrI\(_3\) remains centrosymmetric, regardless of any rigid translation between the two sheets. Therefore, i-type SHG in bilayer CrI\(_3\) is forbidden under the electric dipole approximation. On the other hand, c-type SHG originating from the magnetic structure could arise owing to the layered antiferromagnetic order\(^7\). As shown in Fig. 1c, the two-layered antiferromagnetic configurations, with all spins pointing outwards or inwards, break both time-reversal and spatial-inversion symmetries, allowing electric-dipole c-type SHG. By contrast, when the bilayer is driven into fully spin-aligned states upon the application of an out-of-plane magnetic field, the inversion symmetry of the magnetic structure is restored (Fig. 1d) and prohibits c-type SHG. Thus, these unique magnetic structures in bilayer CrI\(_3\) can be tuned, allowing the exploration of magnetization-induced electric-dipole SHG in the atomically thin limit, which in turn may reveal subtle structural information associated with interlayer magnetic coupling that cannot be readily obtained with existing approaches.

We first investigate layered antiferromagnetism-induced nonreciprocal SHG in CrI\(_3\) bilayers as a function of temperature and out-of-plane magnetic field. Unless otherwise noted, a pulsed 900-nm femtosecond laser at a power of 0.6 mW was used (see details in Methods). CrI\(_3\) bilayers were mechanically exfoliated and encapsulated by thin hexagonal boron nitride (hBN) flakes to prevent degradation\(^7\). Figure 1e shows an optical microscope image of the sample (white dashed outline) used to obtain the data reported here, before hBN encapsulation; also shown in a neighbouring thicker flake. Figure 1f–h shows SHG microscope images of the same area. At 50 K, above the critical temperature (about 40 K) of bilayer CrI\(_3\), no SHG signal is observed (Fig. 1f). When the sample was cooled to 5 K at zero magnetic field, a layered antiferromagnetic state forms and a strong and homogenous SHG signal emerges (Fig. 1g). The signal from the bilayer vanishes at a magnetic field of ~1 T (Fig. 1h), which aligns all spins. The isolated thicker CrI\(_3\) flake has a slightly different SHG response compared to the bilayer, owing to its increased thickness and different magnetic structure (Extended Data Fig. 1), which is not the focus of this work. For comparison, we also studied a monolayer of CrI\(_3\), in which no SHG signal was observed from either its lattice or its magnetic structure (Extended Data Fig. 2), consistent with the symmetry analysis above. The SHG signal from thin hBN flakes used for encapsulation is barely observable, because the bulk crystal of hBN is centrosymmetric\(^12\).

We extract the SHG intensity of the bilayer at zero magnetic field and plot it as a function of temperature in Fig. 1i. The SHG signal depends sensitively on the temperature and vanishes above the critical temperature, where the layered antiferromagnetic order disappears. Similar behaviour was observed in other CrI\(_3\) bilayers, as shown in Extended Data Fig. 3. The vanishing SHG above the critical temperature, as well as
in the fully spin-aligned states, also confirms the centrosymmetric lattice structure of CrI_3 bilayers. The combined magnetic-field-dependent and temperature-dependent measurements unambiguously point to layered antiferromagnetic order as the origin of the SHG.

This nonreciprocal c-type SHG enables us to probe the structural symmetry of layered antiferromagnetic states in bilayer CrI_3, for which conventional means—including neutron diffraction and the magneto-optical Kerr effect—are difficult to apply\(^6\). Such information is currently lacking, but is crucial for understanding the microscopic origin of the antiferromagnetic interlayer coupling in bilayer CrI_3. In particular, whereas bulk CrI_3 crystals are known to be ferromagnetic\(^{14}\), CrI_3 bilayers exhibit layered antiferromagnetism. Recent theoretical calculations\(^{15–18}\) have shown that the magnetic interlayer coupling in CrI_3 depends critically on the stacking structure, that is, how the two monolayer sheets in the bilayer are laterally coupled with respect to each other. In particular, the rhombohedral stacking structure is predicted to favour ferromagnetic interlayer coupling, whereas the monoclinic stacking structure is antiferromagnetically coupled. On the other hand, it has been reported that bulk CrI_3 crystal has a rhombohedral structure at low temperature and a monoclinic structure at high temperature, with a structural phase transition\(^{14}\) occurring at around 200 K. Thus, the stacking structure, or magnetic structure due to spin–lattice coupling, in bilayer CrI_3 at low temperatures is an open question.

As illustrated in Extended Data Fig. 4, the rhombohedral structure belongs to the \(S_h\) crystallographic point group, which has an out-of-plane \(C_3\) axis and lacks a mirror plane. By contrast, the monoclinic structure possesses \(C_{2v}\) symmetry, which has an in-plane \(C_2\) axis and a mirror plane. Polarization-resolved SHG measurements are ideally suited to distinguish the symmetry difference between the two stacking structures. Briefly, for a system with three-fold rotational symmetry, because of angular momentum conservation associated with the photon's helicity\(^{26}\), the SHG process should have a cross-circularly polarized optical selection rule, as depicted in Fig. 2a. That is, the absorption of two \(\sigma^+\) photons at the fundamental frequency leads to the emission of one \(\sigma^-\) photon at twice the fundamental frequency, and vice versa, as demonstrated in monolayer transition-metal dichalcogenides\(^{30,31}\). However, although our measurements at zero applied magnetic field show that cross-circularly polarized SHG is indeed the strongest component in the layered antiferromagnetic state (Fig. 2b, c), the SHG signals for co-circularly polarized excitation and detection are appreciable (Fig. 2d, e). Figure 2f shows the corresponding SHG spectra obtained from co- and cross-polarized measurements. These observations suggest the lack of three-fold rotational symmetry in the spin–lattice structure of CrI_3, that is, CrI_3 bilayers do not have a rhombohedral stacking structure at low temperatures.

This was further confirmed by polarization-dependent azimuthal SHG measurements in the linear-polarization basis. In these measurements, the excitation and detection beams were linearly co- (XX) or cross- (XY) polarized while rotating together with respect to the sample plane. Figure 2g shows the polarization dependence under 900-nm excitation. Both XX (black dots) and XY (red dots) patterns show six asymmetric lobes, confirming the broken three-fold rotational symmetry. In fact, the XX and XY patterns are nicely fitted by the solid curves obtained from the c-type second-order nonlinear tensors associated with \(C_{2v}\), crystallographic symmetry, which corresponds to the monoclinic stacking structure (see details in Methods). These observations and fits are robust for different fundamental-wavelength excitations. Figure 2h, i shows the azimuthal-polarization dependence of the SHG signal at 970-nm and 1,040-nm excitation. Although the details of the SHG patterns vary with excitation wavelength, the revealed \(C_{2v}\) symmetry is consistent with an in-plane \(C_2\) axis about 145° with respect to the horizontal direction in Fig. 1e. The details of the SHG polarization dependence are probably determined by the electronic states at the excitation energy\(^{32}\), which reflect the combined spin, rotation and inversion symmetries of the system. Our SHG study thus provides experimental evidence of monoclinic structure at low temperatures, which is consistent with theoretical predictions\(^{15–18}\) indicating that the monoclinic stacking structure causes the layered antiferromagnetism in bilayer CrI_3.

We further studied the polarization-resolved SHG associated with different magnetic states. Figure 3a shows the SHG intensity as a function of magnetic field under \(\sigma^+\) excitation and \(\sigma^-\) detection (\(\sigma^+ / \sigma^-\)). For comparison, Fig. 3b shows the corresponding magnetic-field-dependent reflectance magneto-circular dichroism (RMCD).
dependence at 0 T, measured with a fundamental wavelength of 900 nm – g SHG spectra. a.u., arbitrary units.

Probing of magnetic switching and domains.

Fig. 3 is considerably larger when the magnetic field is swept upwards from antiferromagnetic states. The data in Fig. 3 show that the SHG signal is non-zero in the layered ferromagnetic states and vanishes in the layered antiferromagnetic state. The SHG signal, by contrast, is non-zero in the layered antiferromagnetic state.

RMCD is non-zero only when the bilayer is in one of the fully spin-aligned ferromagnetic states. Consistent with previous reports, RMCD is non-zero only when the bilayer is in one of the fully spin-aligned ferromagnetic states and vanishes in the layered antiferromagnetic state. The SHG signal, by contrast, is non-zero in the layered antiferromagnetic state.

measurement of the same bilayer. The excitation powers of the 900-nm, 970-nm and 1,040-nm beams were 0.6 mW, 1.0 mW and 0.8 mW, respectively; the latter two powers are increased owing to the weaker SHG signal. The data in f–i were obtained at the position marked by the green dot in Fig. 1g. Solid lines are fits by the c-type second-order nonlinear tensors associated with $C_{2h}$ symmetry (monoclinic stacking structure), as described in Methods.

were linearly polarized, with XX and XY indicating co- and cross-linearly polarized beams, respectively. The azimuthal angle of $0^\circ$ refers to excitation polarization parallel to the horizontal direction in Fig. 1e. The excitation powers of the 900-nm, 970-nm and 1,040-nm beams were 0.6 mW, 1.0 mW and 0.8 mW, respectively; the latter two powers are increased owing to the weaker SHG signal. The data in f–i were obtained at the position marked by the green dot in Fig. 1g. Solid lines are fits by the c-type second-order nonlinear tensors associated with $C_{2h}$ symmetry (monoclinic stacking structure), as described in Methods.

Fig. 2 | Polarization-resolved SHG in the layered antiferromagnetic state. a, Optical selection rules with three-fold rotational symmetry for circularly polarized SHG. The upwards and downwards arrows represent the fundamental and second-harmonic light, respectively. b–e, Polarization-resolved SHG intensity images at zero magnetic field: $\sigma^+ / \sigma^-$ (b), $\sigma^+ / \sigma^-$ (c), $\sigma^- / \sigma^-$ (d) and $\sigma^- / \sigma^-$ (e), f, Polarization-resolved SHG spectra. a.u., arbitrary units. g–i, Azimuthal SHG polarization dependence at 0 T, measured with a fundamental wavelength of 900 nm (g), 970 nm (h) and 1,040 nm (i). The excitation and detection beams were linearly polarized, with XX and XY indicating co- and cross-linearly polarized beams, respectively. The azimuthal angle of $0^\circ$ refers to excitation polarization parallel to the horizontal direction in Fig. 1e. The excitation powers of the 900-nm, 970-nm and 1,040-nm beams were 0.6 mW, 1.0 mW and 0.8 mW, respectively; the latter two powers are increased owing to the weaker SHG signal. The data in f–i were obtained at the position marked by the green dot in Fig. 1g. Solid lines are fits by the c-type second-order nonlinear tensors associated with $C_{2h}$ symmetry (monoclinic stacking structure), as described in Methods.

—1 T than when the field is swept downwards from 1 T. If the polarization setting is switched to $\sigma^- / \sigma^-$—equivalent to applying a time-reversal operation on the spin–lattice structure—the intensities are reversed (Extended Data Fig. 5). This distinct difference in the SHG signal between the two field-sweep directions demonstrates the existence of two antiferromagnetic ground states, as schematically shown in Fig. 3c.

Fig. 3 | Probing of magnetic switching and domains. a, Circularly polarized SHG intensity as a function of magnetic field. The excitation is $\sigma^+$-polarized and the detection is $\sigma^-$-polarized. The data were taken at the sample position marked by the green dot in Fig. 1g. The magnetic field was swept downwards (red) and upwards (blue). b, Corresponding RMCD hysteresis loop. c, Schematic of the evolution of magnetic states driven by the magnetic field. d–h, Circularly polarized SHG intensity, obtained with the $\sigma^- / \sigma^-$ configuration at selected magnetic fields near the metamagnetic transition, while the field was swept upwards. The images reveal domain effects in the layered antiferromagnetic states.
Thus, the two antiferromagnetic ground states can be independently studied, as shown in Fig. 2 and Extended Data Fig. 6.

In addition, SHG can also probe magnetic-domain dynamics near the metamagnetic transition. Figure 3d-h shows SHG microscope images at selected magnetic fields. At $-0.63$ T, no SHG signal is observed in the CrI$_3$ bilayer, implying that the whole bilayer is in the fully spin-aligned state. As the field is swept to $-0.58$ T, the lower half of the CrI$_3$ bilayer displays a SHG signal, corresponding to domain switching from the spin-aligned state to the layered antiferromagnetic state. As the field is swept continuously to $-0.52$ T, the SHG signal in the upper half of the CrI$_3$ bilayer gradually appears in spots, as more domains switch from the spin-aligned state to the layered antiferromagnetic state. We note that the domain switching observed by magneto-optical Kerr effect microscopy is different, because the magneto-optical Kerr effect is sensitive to ferromagnetic states. With its sensitivity to the layered antiferromagnetic states, SHG microscopy provides a powerful technique to image antiferromagnetic domain switching in CrI$_3$ bilayers.

Lastly, we discuss the strength of the layered antiferromagnetism-induced SHG. Given the Cs$_3$ lattice symmetry, we estimate the second-order nonlinear susceptibilities $\chi^{(2)}_{xx}$ and $\chi^{(2)}_{xyy}$ following the procedure detailed in Methods. Because magnetization-induced SHG is generally weak, the estimated c-type $\chi^{(2)}_{xyy}$ is $2 \text{ nm}^{-1}$ of the CrI$_3$ bilayer is remarkably strong (Extended Data Fig. 7). For a direct comparison, we measured the SHG signal from a bulk Cr$_2$O$_3$ crystal with a (0001) surface (Extended Data Fig. 8), a model system for studying antiferromagnetism-induced SHG. In Cr$_2$O$_3$ bulk, the electric-dipole-allowed c-type SHG is comparable to the magnetic dipole contribution of i-type SHG from the lattice, as shown in Fig. 4, the SHG intensity at the two-photon resonant excitation (580 nm) is about three orders of magnitude smaller than that for bilayer CrI$_3$. Given the coherence length of bulk Cr$_2$O$_3$, the estimated value of the c-type $\chi^{(2)}_{xyy}$ is about 2 pm V$^{-1}$, which is three orders of magnitude weaker than that in bilayer CrI$_3$. Additionally, it is also known that electric-dipole-allowed c-type SHG is present at the surface or interface of ferromagnetic transition-metal thin films, where both spatial-inversion and time-reversal symmetries are broken. The corresponding magnitude of this c-type $\chi^{(2)}_{xyy}$ is substantially lower, of the order of $10^{-7}$ pm V$^{-1}$. Negligible SHG in the CrI$_3$ bilayer and monolayer with fully aligned spin states corroborate this weak surface effect.

We also measured the SHG responses in monolayers of MoS$_2$ and hBN using the same experimental setup (Fig. 4, Extended Data Fig. 9). Here, MoS$_2$ monolayers were exfoliated onto SiO$_2$/Si substrates, whereas hBN monolayers were grown by chemical vapour deposition and transferred onto a SiO$_2$/Si substrate. The $\chi^{(2)}$ value of CrI$_3$ bilayers is about five times higher than that of hBN monolayers and is comparable to that of MoS$_2$ monolayers at the two-photon resonance with the 1s exciton (660 nm). Because monolayer MoS$_2$ exhibits the highest $\chi^{(2)}$ value amongst all known 2D materials, which is comparable to those of the best-known second-order nonlinear crystals, the discovery of strong c-type SHG from the layered antiferromagnetism in bilayer CrI$_3$ is extraordinary. Our results therefore highlight the opportunities of applying SHG to investigate 2D antiferromagnetic order, as well as exploring nonreciprocal nonlinear optics with possible control at the atomically thin limit.

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when 90 or isponding magnetic point group is of-plane layered antiferromagnetic spin configuration into account, the corre-rotation operator with an in-plane axis, monoclinic stacking structure in a CrI3 bilayer, which has the crystallographic zation dependence measurements and magnetic hysteresis loops were obtained and detection beams with a quarter- or half-wave plate, in combination with linear or a spectrograph equipped with a liquid-nitrogen-cooled charge-coupled device. The reflected SHG signal was detected in photon-counting mode and its power was calibrated by a beam of known power at 2ω propagating along the same path to the same detector. Given the thickness of bilayer CrI3 of 1.4 nm, we obtained [x|x] ≈ 1.1 nm V−1 and [x|y] ≈ 1.7 nm V−1 (the wavelength dependence of [x|2] is shown in Extended Data Fig. 7). Similarly, the [x|2] values of monolayer MoS2 and monolayer hBN could be estimated, for which the crystallographic symmetry becomes D3h. In this case, there is only one independent non-zero tensor element: χ3x−,−y− = χ3x−−−−−y−−−−−. Given their different monolayer thicknesses and refractive indices, the [x|2] value of monolayer MoS2 at the 1s exciton two-photon resonance (660 nm) is about 2 nm V−1 and the [x|2] value of monolayer hBN at an excitation wavelength of 900 nm is about 0.4 nm V−1. For bulk Cr2O3 crystal, the effective thickness is estimated by the coherence length (about 73 nm). Thus, the [x|2] value of bulk Cr2O3 crystal at the two-photon resonant transition (580 nm) is approximately 2 pm V−1, consistent with the literature.44

Data availability The authors declare no competing interests.

Additional information Correspondence and requests for materials should be addressed to X.X. or S.W. Peer review information Nature thanks Roman Pisarev, Joaquín Fernández Rosicki and the other, anonymous, reviewer(s) for their contribution to the peer review of this work. Reprints and permissions information is available at http://www.nature.com/reprints.
Extended Data Fig. 1 | SHG intensity images at higher magnetic fields (±2 T). Compared with the SHG intensity images in Fig. 1, no SHG is observed for the CrI\textsubscript{3} bilayer (left in the images) and the SHG is weaker for the thicker flake (right in the images). kcps, 1,000 c.p.s.
Extended Data Fig. 2 | Negligible SHG in a CrI₃ monolayer. a, Optical microscope image of a CrI₃ monolayer (delineated by the white dashed line). b–e, Corresponding SHG intensity images when the monolayer is nonmagnetic (b; 0 T, 50 K), ferromagnetic (c–e; 0 T and ±1 T, 5 K). f, Corresponding RMCD hysteresis loop at 5 K, showing the appearance of ferromagnetism in the monolayer.
Extended Data Fig. 3 | SHG intensity as a function of temperature for a different CrI₃ bilayer sample. The bilayer was in a layered antiferromagnetic state with no external magnetic field applied. The red solid curve is a guide for the eye following the power law $\left| 1 - \frac{T}{T_c} \right|^2$ when $T < T_c$. 
Extended Data Fig. 4 | Possible stacking structures in bilayer CrI$_3$ with distinct crystallographic symmetry. 

**a.** Atomic structure of monolayer CrI$_3$, as in Fig. 1a. 

**b, c.** Rhombohedral (b) and monoclinic (c) stacking structures in bilayer CrI$_3$. The rhombohedral structure belongs to the $S_6$ crystallographic point group, which has an out-of-plane $C_3$ axis and lacks a mirror plane. By contrast, the monoclinic structure has $C_{2h}$ symmetry, which has an in-plane $C_2$ axis and a mirror plane. We note that if the monolayer sheets are laterally translated along the mirror plane in the monoclinic stacking structure, $C_{2h}$ symmetry remains.

**d.** Side view of the monoclinic stacking structure.
Extended Data Fig. 5 | Circularly polarized SHG intensity as a function of magnetic field. The excitation is $\sigma^-$-polarized and the detection is $\sigma^+$-polarized. The data were taken at the sample position marked by the green dot in Fig. 1g.
Extended Data Fig. 6 | Polarization-resolved SHG in the other layered antiferromagnetic state. a–d, Polarization-resolved SHG intensity images at zero magnetic field: $\sigma^+/\sigma^-$ (a), $\sigma^-/\sigma^+$ (b), $\sigma^+/\sigma^+$ (c) and $\sigma^-/\sigma^-$ (d). Here the magnetic field was swept upwards from $-1 \text{ T}$ to $0 \text{ T}$, in contrast to the downwards sweeping direction shown in Fig. 2. e, Corresponding polarization-resolved SHG spectra. f–h, Azimuthal SHG polarization dependence at $0 \text{ T}$ with a fundamental wavelength of $900 \text{ nm}$ (f), $970 \text{ nm}$ (g) and $1,040 \text{ nm}$ (h). The excitation and detection beams were linearly polarized, with XX and XY polarization. The azimuthal angle of $0^\circ$ refers to an excitation polarization parallel to the horizontal direction in Fig. 1c. The excitation powers of the $900$-nm, $970$-nm and $1,040$-nm beams were $0.6 \text{ mW}$, $1.0 \text{ mW}$ and $0.8 \text{ mW}$, respectively. Data in e–h were obtained at the position marked by the green dot in Fig. 1g. Solid lines are fits by the c-type second-order nonlinear tensors associated with $C_{3h}$ symmetry (monoclinic stacking structure), as described in Methods.
Extended Data Fig. 7 | Extracted second-order susceptibilities $|\chi^{(2)}|$ of a CrI$_3$ bilayer. The sample was at 5 K, without an applied magnetic field. The excitation wavelength was varied from 800 nm to 1,040 nm. The method used to extract the second-order susceptibilities is detailed in Methods.
Extended Data Fig. 8 | SHG study of a bulk Cr₂O₃ crystal at different excitation wavelengths and powers.  

**a**, SHG spectra, with the fundamental wavelength tuned from 1,000 nm to 1,300 nm. The excitation power was 10 mW and the sample temperature was 8 K. The excitation beam was linearly polarized, and no polarization analyser was used for detection. The variation in SHG intensity reflects the resonant electronic transitions in bulk Cr₂O₃ crystals, consistent with the original report by Fiebig et al.¹⁹.  

**b**, Dependence of SHG intensity on the excitation power. On this log–log scale plot, the data (open circles) are fitted linearly with a slope of $\alpha = 2.03 \pm 0.16$ (dashed line), confirming the quadratic power dependence of SHG. The excitation wavelength is 1,160 nm.
Extended Data Fig. 9 | Comparison between the second-order nonlinear susceptibility of different 2D materials. The open squares show the measured second-order susceptibilities of monolayer MoS$_2$ (red), monolayer hBN (purple) and bilayer CrI$_3$ in the antiferromagnetic state (orange). The measurement was conducted using the same set-up (Extended Data Fig. 10), with a second-harmonic photon energy of 2.76 eV (wavelength, 450 nm). The red curve shows the $|\chi^{(2)}|$ spectrum of monolayer MoS$_2$, with the second-harmonic photon energy ranging from 1.8 eV to 3.2 eV.
Extended Data Fig. 10 | Optical layout used for the SHG measurement. GM, two-axis galvanometer; M, silver mirror; SL, scan lens; P1 and P2, Glan–Thompson polarizers; BS, beamsplitter; TL, tube lens; WP, wave plate; SCM, superconducting magnet; FM, flip mirror; PMT, photomultiplier tube in photon-counting mode; and L, focusing lens.