Tuning inelastic light scattering via symmetry control in the two-dimensional magnet CrI$_3$

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The coupling between spin and charge degrees of freedom in a crystal gives rise to magneto-optical effects with applications in the sensitive detection of local magnetic order, optical modulation and data storage. In two-dimensional magnets these effects manifest themselves in the large magneto-optical Kerr effect$^{2,3}$, spontaneous helical light emission$^{4-6}$ from ferromagnetic (FM) monolayers and electric-field induced Kerr rotation$^{7-9}$ and giant second-order non-reciprocal optical effects$^{6}$ in antiferromagnetic (AFM) bilayers. Here we demonstrate the tuning of inelastically scattered light through symmetry control in atomically thin chromium triiodide (CrI$_3$). In monolayers, we found an extraordinarily large magneto-optical Raman effect from an A$_{1g}$ phonon mode due to the emergence of FM order. The linearly polarized, inelastically scattered light rotates by $\sim$40°, more than two orders of magnitude larger than the rotation from the magneto-optical Kerr effect under fluctuations of FM order. The linearly polarized, inelastically scattered light from the even-parity phonon mode. Our work demonstrates the magneto-electrical control over these selection rules by activating or suppressing Raman activity for the odd-parity phonon mode and the magneto-optical rotation of scattered light from the even-parity phonon mode. Our work underlines the unique opportunities provided by two-dimensional magnets to control the combined time-reversal and inversion symmetries to manipulate Raman optical selection rules and for exploring emergent magneto-optical effects and spin-phonon coupled physics.

Raman scattering measures light inelastically scattered from collective quasiparticle excitations. As it is highly sensitive to material parameters, such as crystal symmetry and local electronic states, Raman spectroscopy provides a powerful probe of a broad range of condensed-matter phenomena, such as charge density waves$^6$, superconductivity$^{10}$, ferroelectricity$^{11}$ and topological physics$^{12}$. In particular, Raman scattering from spin–phonon excitations has yielded incisive information on magnetic materials. For instance, in recently developed two-dimensional van der Waals magnets, Raman scattering was used to reveal magnetic order and phase transitions$^{13-15}$ down to a single layer$^{16-18}$.

CrI$_3$, a van der Waals magnet, was shown to be a layered antiferromagnet in its few-layer form: spins within each layer are ferromagnetically coupled with strong out-of-plane anisotropy, whereas the interlayer exchange is AFM$^1$. In a bilayer, on the application of a moderate magnetic field, the system undergoes a spin–flip transition, switching from a layered AFM state to a fully spin-polarized state. Second harmonic generation measurements have revealed the restoration of inversion symmetry when the bilayer is switched from the AFM state to the fully spin-polarized state, which highlights the dependence of symmetry on the magnetic order in CrI$_3$ bilayers. As such, Raman optical selection rules, and hence specific Raman modes, are likely to be controllable when switching between magnetic states. Given the predicted and recently reported strong spin–lattice coupling in the monolayer$^{19,20}$, atomically thin CrI$_3$ is a promising candidate to study tunable magneto-optical Raman effects in the two-dimensional limit.

In this work, atomically thin CrI$_3$ flakes were mechanically exfoliated onto oxidized silicon substrates and sandwiched between two flakes of hexagonal boron nitride to prevent their degradation in air. These samples were cooled in a cold-finger cryostat and excited at the normal incidence using a HeNe laser at a wavelength of 632.8 nm, nearly resonant with the ligand-to-metal charge-transfer transition near 2 eV (ref. 3) (Methods gives the sample fabrication and measurement details).

We first present the Raman scattering results from monolayers of CrI$_3$. At 60 K, above the Curie temperature ($T_C$) of $\sim$45 K, the monolayer is paramagnetic, as confirmed by the reflective magneto circular dichroism (RMCD) measurements shown in the inset of Fig. 1a. Raman spectra taken at this temperature in the co- and cross-linear polarization channels, which we denote XX and XY, respectively, show four distinct peaks (Fig. 1a). Based on their observed polarization dependence as well as the calculated phonon modes$^{22}$, we assigned the peaks at 76.9 cm$^{-1}$ and 127.4 cm$^{-1}$ as scattering from A$_{1g}$ phonons and the peaks at 107.7 cm$^{-1}$ and 114.8 cm$^{-1}$ as scattering from two of the four E$_g$ phonons. The other two predicted E$_g$ phonons at around 50 cm$^{-1}$ and 230 cm$^{-1}$ were not seen, possibly due to their weak scattering cross-section. We note that there is residual Raman scattering in the XY channel from the 127.4 cm$^{-1}$ A$_{1g}$ phonon that is absent from the 76.9 cm$^{-1}$ A$_{1g}$ phonon. Such Raman activity in the XY channel is typically forbidden for A$_{1g}$ phonons, but can be enabled for certain phonon modes through mechanisms such as resonant effects$^{22}$.

When cooled below $T_C$, the monolayer becomes FM, evident by the magnetic hysteresis in the RMCD measurements (Fig. 1b, left inset). As seen in Fig. 1b, there is a significant increase in the cross-linearly polarized Raman scattering from the 127.4 cm$^{-1}$ A$_{1g}$ phonon (Supplementary Fig. 1 gives data in the opposite spin orientation). The other A$_{1g}$ phonon at 76.9 cm$^{-1}$ and the two E$_g$ phonons...
show no marked change in their Raman signal in both the XX and the XY channels. As such, for the rest of our discussion on the CrI₃ monolayer, we focus on only the 127.4 cm⁻¹ A¹g phonon, which corresponds to an out-of-plane and out-of-phase vibration between the two iodine layers (Fig. 1b, right inset).

To understand the sudden enhancement of the 127.4 cm⁻¹ peak in the XY channel, we tracked its intensity while changing the relative angle of the analysing polarizer with respect to a fixed laser excitation polarization (green double-sided arrow along the 0°–180° axis in Fig. 2a,b (Methods)). Above Tᶜ, the resultant polarization pattern at 60 K in Fig. 2a shows that this phonon mode exhibits A¹g symmetry—the polarization axis, delineated by a dotted line, is co-linear with the laser excitation. In stark contrast, the polarization axis in the FM state at 15 K is rotated by some angle, φ₁ away from the excitation polarization (Fig. 2b). The rotation is roughly equal (about 40°) and opposite between the two time-reversal paired FM ground states, does not depend on the laser excitation polarization (Supplementary Fig. 2). In general, time-reversal symmetry breaking can induce a Hall-like, antisymmetric component in the Raman tensor of an A¹g phonon (Methods) that is responsible for the polarization rotation of the scattered light. However, what is unexpected is that this rotation by a monolayer FM insulator is remarkably large, being two orders of magnitude larger than that from the magneto-optical Kerr effect. A similar, albeit smaller, rotation can also be observed in the 76.9 cm⁻¹ peak, shown in Supplementary Fig. 3.

We also performed temperature-dependent measurements of the polarization pattern, initially at 60 K and cooling through Tᶜ without applying a magnetic field (Supplementary Fig. 4a–f). Figure 2c shows the extracted polarization rotation, φ₁, at selected temperatures down to 15 K. φ₁ has an abrupt onset precisely when the FM order is established at around 45 K and increases as the sample is further cooled down to 15 K. This temperature-dependent evolution of φ₁ matches the emergence of ferromagnetism seen in the remnant RMCD signal (Supplementary Fig. 4g). Combined with the fact that φ₁ was equal and opposite between the two FM states, we conclude that the origin of the polarization rotation is from the FM order in the monolayer.

Switching to the circular polarization basis further reveals the effects of FM order on the Raman scattering from the 127.4 cm⁻¹ A¹g phonon. Figure 2d–f shows the helicity-resolved Raman scattering measurements from the 127.4 cm⁻¹ mode for the four possible scattering channels, σ⁺/σ⁺, σ⁺/σ⁻, σ⁻/σ⁺ and σ⁻/σ⁻, of which the first describes the incident helicity and the last describes the outgoing helicity (the σ⁺/σ⁻ and σ⁻/σ⁺ scattering is schematically drawn in the insets of Fig. 2d). In Fig. 2d, Raman scattering measurements above Tᶜ at 60 K show equal scattering in the σ⁺/σ⁺ and σ⁻/σ⁺ channels, in which the outgoing helicity is preserved. There is negligible scattering in the σ⁺/σ⁻ and σ⁻/σ⁺ channels, in which the outgoing helicity is reversed. In stark contrast, below Tᶜ at 15 K, Fig. 2e shows that, with the magnetization pointing up, the 127.4 cm⁻¹ mode is dominated by the σ⁺/σ⁺ channel. By flipping the magnetization, the Raman scattering is then dominated by the σ⁻/σ⁻ channel (Fig. 2f), the exact time-reversed process of that observed when the magnetization pointed up.

Next, we explored the effects of magnetic order on the magneto-optical Raman scattering from bilayers of CrI₃. Unless stated otherwise, all the following measurements were performed at 15 K. Unlike the monolayer, at zero magnetic field two peaks distinct in energy appeared at 126.7 cm⁻¹ and 128.8 cm⁻¹ in the XY and XX channels, respectively (Fig. 3a). When the field was above the spin–flip transition (0.7 T) to fully align the spins, only a single peak was observed at 128.8 cm⁻¹ in both the XX and XY channels (Fig. 3b). Figure 3c–e shows the Raman intensity of these channels plotted as a function of magnetic field and Raman shift, whereas the magnetic field-dependent RMCD signal, shown in Fig. 3d, provides information on the corresponding magnetic states.

Starting with the XX channel, the mode at 128.8 cm⁻¹ does not change in either energy or intensity as the magnetic field varies (Fig. 3e). In contrast, the XY channel in Fig. 3c shows that the 126.7 cm⁻¹ peak present in the AFM states is abruptly suppressed when the bilayer is switched to the fully spin-polarized states. Simultaneously, a peak at 128.8 cm⁻¹ emerges in the cross-polarized channel. The temperature dependence of the peaks in the XY channel further confirms their magnetic origin. Above Tₓ≈45 K in the absence of an applied magnetic field, we only observed a single co-linearly polarized peak at 128.1 cm⁻¹, which slightly blue-shiftings to 128.8 cm⁻¹ as the bilayer is cooled to 15 K (Fig. 3g). Below Tₓ, both cross-polarized peaks at 126.7 cm⁻¹ (AFM state) and 128.8 cm⁻¹ (fully spin-polarized state) appear with the onset of magnetic order (Fig. 3f and Supplementary Fig. 5).

The appearance of the two Raman peaks in the AFM state and the dependence of the cross-linearly polarized spectra on the magnetic order can be understood by treating the CrI₃ bilayer as a coupled spring system. In a bilayer, weak van der Waals interactions between the two layers split each phonon mode in the monolayer into two modes (that is, Davydov splitting). Given that the lattice of bilayer CrI₃ is centrosymmetric, we can classify one of the two modes as an odd-parity mode (u) and the other as an even-parity mode (g). For the 127.4 cm⁻¹ A¹g mode in the monolayer, Davydov splitting in the bilayer results in a lower-energy 126.7 cm⁻¹ u mode, in which the layers vibrate out-of-phase, as depicted in Fig. 3h,
and a higher-energy 128.8 cm\(^{-1}\) g mode with in-phase vibrations between the layers (Fig. 3i). As Raman scattering is an even-parity process, this implies that the even-parity 128.8 cm\(^{-1}\) g mode is Raman active, whereas the odd-parity 126.7 cm\(^{-1}\) u mode is infrared active but Raman silent. This is consistent with the Raman spectra above \(T_c\) (Fig. 3g).

Factoring in magnetic order leads to remarkable changes in the Raman optical selection rules. For the fully spin-polarized state, the centrosymmetry remains intact (Fig. 3i), so it should behave exactly like the FM monolayers: the 128.8 cm\(^{-1}\) g mode is active in both the XX and XY channels, whereas the 126.7 cm\(^{-1}\) u mode remains silent. This is consistent with experimental findings. A comparison of the linear polarization patterns between the 127.4 cm\(^{-1}\) A\(_{1g}\) phonon mode in the FM monolayer and the 128.8 cm\(^{-1}\) g mode in the bilayer shows virtually the same degree of polarization rotation (Supplementary Fig. 6). In the AFM state, parity no longer applies because the antiparallel spin configuration breaks the inversion symmetry (Fig. 3k). Yet, the system remains invariant under the combined time-reversal and inversion symmetry, which forbids Raman activity in the Hall-like component in the Raman tensor of the bilayer monolayer are not visibly affected by magnetic order, which implies that the terms related to the magnetic order are negligibly small or do not couple to the excitation based on the scattering geometry. These additional terms, however, enable the Raman activity of the odd Davydov-split A\(_{1g}\) modes at 126.7 cm\(^{-1}\) and 76.4 cm\(^{-1}\). Thus, as the terms related to the magnetic order in the E\(_{g}\) Raman tensors are negligibly small, Raman activity of the odd Davydov-split E\(_{g}\) mode will not be detectable through Raman scattering measurements.

Last, we demonstrated magneto-electrical control over the Davydov-split phonons by electrically switching the magnetic states of the bilayer device shown in Fig. 4a,b. The magnetic-field-dependent Raman intensity plot taken near the spin-flip transition at an applied gate voltage, \(V_g\), of 0 V (5 V) is shown in Fig. 4c,d. The magnetic field at which the 126.7 cm\(^{-1}\) peak is activated or suppressed is modulated from −0.7 to −0.6 T, consistent with previous electrical control of the spin-flip transition\(^{6}\). With the magnetic field at −0.62 T, Fig. 4e,f compares the co- and cross-linearly polarized Raman spectra at two different \(V_g\) values, 0 and 5 V. At 5 V, the 126.7 cm\(^{-1}\) peak is suppressed, whereas the 128.8 cm\(^{-1}\) peak is activated in the XY channel as the positive \(V_g\) switches the magnetic states. For Fig. 4g, we continuously swept \(V_g\) from −1 V up to 6 V and monitored the Raman activity of the 126.7 cm\(^{-1}\) and 128.8 cm\(^{-1}\) phonons in the XY channel. The gradual suppression of the 126.7 cm\(^{-1}\) phonon and emergence of the 128.8 cm\(^{-1}\) phonon track exactly the gate-dependent RMCD signal (Fig. 4h) with a progressively larger negative RMCD signal as the bilayer was switched from an AFM state to the fully spin-down polarized state. From these measurements, we confirmed the magneto-electrical switching of the 126.7 cm\(^{-1}\) phonon mode through electrostatic control of the magnetic states, and hence the Raman selection rules, in a gated CrI\(_3\) bilayer device.

In summary, we observed a giant rotation (~40°) of linearly polarized inelastically scattered light by a monolayer FM insulator, established to originate from its magnetic order. Although we
Fig. 3 | Coupling of magnetic order and Raman optical selection rules in bilayer CrI₃. a,b, Co- and cross-polarized Raman spectra taken in an AFM state at a zero applied magnetic field (a) and the fully spin-up polarized state at 1.5 T with a zero V<sub>g</sub> (b). c, Colour map of Raman spectra at a range of applied magnetic fields swept from 1.5 to −1.5 T in the cross-linear scattering channel. d, Magnetic field-dependent RMCD signal of the same bilayer as in c. e, Colour map of Raman spectra taken in the co-linear scattering channel. f, Colour maps of cross-linearly polarized (f) and co-linearly polarized (g) Raman spectra taken at a range of temperatures while warming from 15 to 85 K in a zero applied magnetic field. The white dashed line denotes the Néel temperature, T<sub>N</sub>, at which the mode in the cross-linear scattering channel is suppressed. From this, T<sub>N</sub> ≈ 45 K. h,i, Illustrations of the two Davydov-split A<sub>1g</sub> modes in a CrI<sub>3</sub> bilayer: an infrared-active peak at 126.7 cm<sup>−1</sup> (h) and a Raman-active peak at 128.8 cm<sup>−1</sup> (i). j, In the fully spin-polarized states, applying the inversion operation, −r, preserves the spin orientation, and therefore the centrosymmetry of the entire bilayer. k, However, applying −r in the layered AFM states switches the spin orientation of the two layers, and thus breaks centrosymmetry. However, the combined time-reversal and inversion symmetry still holds.

Fig. 4 | Electrical switching of a Raman-silent phonon in bilayer CrI₃. a, Schematic of a gated CrI₃ bilayer device. b, Optical micrograph of the assembled gated CrI₃ bilayer device in a. Scale bar, 5 µm. c,d, Colour map of the Raman spectra at a range of applied magnetic fields swept from −0.4 to −0.8 T in the cross-linear scattering channel of a CrI₃ bilayer device taken at a V<sub>g</sub> of 0 V (c) and 5 V (d). The white dashed line denotes the magnetic field at which the spin–flip transition occurs in the ungated CrI₃ bilayer. e,f, Co- (black) and cross-linearly (red) polarized Raman spectra taken at a V<sub>g</sub> of 0 V (e) and 5 V (f) at a fixed magnetic field of −0.62 T. g, Colour map of cross-linearly polarized Raman spectra taken at range of V<sub>g</sub> values from −1 V to 6 V at a fixed magnetic field of −0.62 T. h, RMCD signal of the same device taken in the same experimental conditions as in g. Gr, graphite; 2L, two layer.
analysed the optical selection rules based on symmetry, a quantitative understanding of the rotation calls for a microscopic theory that considers the resonant Raman excitation. In bilayers of CrI₃, we demonstrated the Raman activation of a symmetry-forbidden mode caused by the emergence of a layered AFM order. This coupling between the Raman selection rules and the combination of inversion symmetry and magnetic structure enabled a demonstration of the magnetoelastic switching of Davydov-split phonon modes. These findings establish atomically thin CrI₃ as a unique platform for exploring externally sensitive magneto-optical effects through the exploitation of symmetries in the two-dimensional limit.

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Methods

Sample preparation. Bulk crystals of CrI₃ were mechanically exfoliated onto 285 nm SiO₂/Si substrates inside a glove box with a N₂ atmosphere. Monolayer and bilayer flakes were identified by optical contrast with respect to the substrate. Once suitable flakes were identified, they were encapsulated with 20–30 nm thick hexagonal boron nitride (hBN) flakes.

Encapsulated CrI₃ samples were prepared inside the glove box through a dry-transfer technique using a poly(bisphenol A carbonate) film stretched over a polydimethylsiloxane cylinder as our stamp². Each flake was picked up and dropped onto a SiO₂/Si substrate in the order: top hBN, CrI₃, flake, bottom hBN.

The gated bilayer samples involved the addition of a 3–5 nm thick graphite flake so that the order of pick up went: hBN top dielectric, graphite contact, bilayer CrI₃, hBN bottom dielectric, and graphite bottom gate. The entire stack was then dropped onto 7 nm/70 nm thick V/Al contacts fabricated using a standard electron-beam lithography technique.

Optical measurements. All the optical measurements were performed in a closed-cycle helium cryostat with a base temperature of 15 K utilizing the backscattering geometry. A superconducting solenoidal magnet was placed around the sample in a Faraday geometry. An objective lens focused 632.8 nm light from a He/Ne laser down to a spot size of about 3 μm onto the sample at normal incidence.

For Raman scattering measurements, the scattered light was dispersed by a Princeton Acton 250i spectrometer using a 1,200 groove mm⁻¹ diffraction grating and detected using a liquid-nitrogen-cooled charge-coupled device. For the monolayer measurements, we used 80 μW of laser power and 5 min integration times, whereas 150 μW and 5 min integrations were used for the CrI₃ bilayers.

For our polarization-dependent measurements, we started in the co-linear (XX) polarization channel and rotated the half-wave plate by 90° continuously until we scanned through a full 360°. To save time, we only scanned through 180° in the temperature-dependent polarization dependence and duplicated this data for polarizations from 180 to 360°.

For the magneto-Raman experiments, the applied magnetic field induced a Faraday rotation for all the light that passed through the optics within the magnet bore. By measuring the rotation of the linearly polarized Rayleigh scattered light at various fields on SiO₂, we determined the Faraday rotation in our system to be −5° T⁻¹. This rotation was compensated for with our polarization optics.

Magnetic materials may exhibit magnetic circular dichroism, which leads to a difference in the amplitude between the reflected and transmitted right-circularly and left-circularly polarized light. When an upper superposition of right circularly and left circularly polarized light, that is, linearly polarized light, is incident on the material, the reflected and transmitted light become elliptically polarized due to the magnetic circular dichroism. In the reflection geometry, this effect is known as RMCD. RMCD measurements were performed using about 2 μW of power from a 632.8 nm He/Ne laser focused to a 3 μm beam spot. The experimental set-up is similar to that used for previous RMCD measurements of the magnetic order in CrI₃, (ref. 5)³.

Weak-coupling model of bilayer CrI₃. The Raman tensor, R, of a monolayer can be decomposed into a spin-independent diagonal part (Rᵣ) and a spin-dependent anti-symmetric part (Rₚ).

\[ R = Rᵣ + Rₚ = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & B \\ 0 & B & 0 \end{pmatrix} + \begin{pmatrix} A & 0 & 0 \\ 0 & C & 0 \\ 0 & 0 & D \end{pmatrix} = \begin{pmatrix} A & C & 0 \\ -C & A & 0 \\ 0 & 0 & B \end{pmatrix} \]

where Rᵣ describes scattering in the XX channel and Rₚ in the XY channel. If the magnetization is flipped, Rᵣ remains the same but Rₚ changes sign. Under the weak-coupling assumption, the Stokes and anti-Stokes components of the total induced dipole moment can be written as \( p(\pm t) = \sum_{i,j} Rᵣᵢ Rᵣᵢ' Q_{ij} E^{\pm t} = \sum_{i,j} Rₚᵢ Rₚᵢ' Q_{ij} E^{\pm t} \), where ij is the layer index, Q is the normal coordinate of the phonon, E is the electric field and \( ω₊ and ω₋ \) are the phonon and phonon frequencies, respectively.

In the AFM state, we have \( Rᵣᵢ = Rᵣᵢ' = Rᵣ \) and \( Rₚᵢ = -Rₚᵢ' = Rₚ \). The normal coordinates are \( Q₋ = -Q₊ = Q \) for the u mode and \( Q₋ = Q₊ = Q \) for the g mode. The total induced dipole moments for the two modes are \( pₓ₋₋ = 2Rᵣ Q₋E^{\pm t} \) and \( pₓ₋₊ = 2Rₚ Q₊E^{\pm t} \). Therefore, the u mode is active only in the XY channel, and the g mode in the XX channel. For the fully spin-polarized state, the key difference from the AFM state is that \( Rᵣᵢ = Rᵣᵢ' = Rᵣ \) and \( Rₚᵢ = -Rₚᵢ' = Rₚ \). This leads to \( pₓ₋₋ = 2Rᵣ Q₋E^{\pm t} \) and \( pₓ₋₊ = 0 \). Consequently, the g mode is active in both the XX and XY channels, but the u mode is silent. This analysis precisely matches our experimental observations.

Symmetry analysis of the Raman tensor. The magnetic point group of monolayer CrI₃ with an out-of-plane magnetization is D₂₀(S₄) = C₃ᵥ + 4C₃ᵥ, where \( θ \) is the time-reversal operator and \( θ' \) is an in-plane rotation axis. In the notation of Shubnikov and Belov⁴, the magnetic point group is 3M. To analyse the symmetry of the Raman tensor of the A₁g mode, we break it down into a symmetric portion, \( Rᵣ \), and an anti-symmetric portion, \( Rₚ \), as shown above. The former is invariant under time-reversal, and thus is independent of the magnetic structure, whereas the latter changes sign on time-reversal and is a consequence of the appearance of a macroscopic magnetization.

CrI₃ bilayers are in the monoclinic phase. This is represented by the symmetry group, \( C₃ᵥ \) = \( \{ e, c₂, i, σ_z \} \), where \( e \) is the identity operator, \( c₂ \) is the two-fold rotation with an in-plane axis, i is the inversion operator and \( σ_z \) is a reflection whose mirror plane is normal to \( c₂ \). The two Davydov-split phonon modes g and u transform under \( C₃ᵥ \) as:

\[ e(g) = |g⟩, e(u) = |u⟩, c₂(g) = |g⟩, c₂(u) = |u⟩, i(g) = |g⟩, i(u) = |u⟩, σ_z(g) = |g⟩, σ_z(u) = |u⟩ \]

Applying the time-reversal operator has no effect on the phonon modes and leaves them unchanged. Considering magnetic structure, however, leads to distinct magnetic point groups between the fully spin-polarized and AFM states. In the fully spin-polarized state, the magnetic point group is \( C₃ᵥ(S₄) = \{ e, i, c₂, iσ_z, h'BhB' \} \). The presence of inversion symmetry (i) renders the u mode silent in both XX and XY channels and the lack of time reversal (θ) allows the Raman tensor for the g mode to develop an antisymmetric component. For the AFM state, its magnetic point group is \( C₃ᵥ(S₄) = \{ e, c₂, iσ_z, h'BhB' \} \). Again, by requiring that the Raman tensor transforms according to the same representation of the corresponding phonon mode, we found that the g mode is silent in the XY channel and the u mode is silent in the XX channel.

Data availability

The datasets generated during and/or analysed during this study are available from the corresponding author upon reasonable request.

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Author contributions

X.X., B.H. and J.C. conceived the experiment. B.H. and J.C. fabricated and characterized the samples, assisted by E.L.R. and T.S. B.H. and J.C. performed the Raman and magnetic circular dichroism measurements. B.H. and J.C. conceived the experiment. B.H. and J.C. fabricated and characterized the samples, assisted by E.L.R. and T.S. B.H. and J.C. performed the Raman and magnetic circular dichroism measurements. B.H., J.C., X.X. and D.X. analysed and interpreted the results. T.T. and K.W. synthesized the hBN crystals. M.A.M. synthesized and characterized the bulk CrI₃ crystals. B.H., J.C., X.X. and D.X. wrote the paper with input from all the authors. All the authors discussed the results.

Competing interests

The authors declare no competing interests.

Additional information

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