Multiwavelength magnetic coding of helical luminescence in ferromagnetic 2D layered CrI\textsubscript{3}

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Highlights
Realizing strong coupling of WGM helical emission and FM orders by SiO\textsubscript{2} cavity
Achieving reversible multiwavelength magnetic coding in the near-infrared region

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Multiwavelength magnetic coding of helical luminescence in ferromagnetic 2D layered CrI$_3$

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SUMMARY

Two-dimensional (2D) van der Waals (vdW) ferromagnets have opened new avenues for manipulating spin at the limits of single or few atomic layers, and for creating unique magneto-exciton devices through the coupling of ferromagnetic (FM) orders and excitons. However, 2D vdW ferromagnets explored so far have rarely possessed exciton behaviors; to date, FM CrI$_3$ have been revealed to show ligand-field photoluminescence correlated with FM ordering, but typically with a broad emission peak. Here, we report a straightforward approach to realize strong coupling of narrow helical emission and FM orders in CrI$_3$ through microsphere cavity. The resonant whispering-gallery modes (WGM) of SiO$_2$ microspheres cause strong oscillation helical emissions with a full width at half-maximum (FWHM) of $\lambda/245$ nm under continuous wave excitation. Reversible magnetic coding of helical luminescence is realized in the range of 950–1100 nm. This work enables numerous opportunities for creating magnetic encoding lasing for photonic integrated chips.

INTRODUCTION

The emergence of long-range ferromagnetic orders in two-dimensional (2D) van der Waals (vdW) ferromagnetic (FM) material has provided a new avenue for creating on-chip lasers, isolators, and modulators for silicon photonics toward information processing and transmission through magneto-exciton coupling (Deng et al., 2018; Gong et al., 2017; Huang et al., 2017; Liu et al., 2020, 2021; Mak et al., 2019; Song et al., 2018; Sun et al., 2019). The magnetic orders can be switched between FM and atomic force microscope (AFM) states by magnetic field (Klein et al., 2018; Zhong et al., 2017), electric gating (Huang et al., 2018; Jiang et al., 2018a, 2018b; Zhang et al., 2020), electrostatic doping (Jiang et al., 2018a; Zheng et al., 2020), and hydrostatic pressure (Li et al., 2019; Song et al., 2019), which empower a pivotal foundation for realizing magnetic and electric control of magneto-optical coupling for information transfer. 2D vdW bilayer CrI$_3$ FM insulators used as spin valve in a magnetic tunneling junction device have highlighted flexibilities for encoding information and data storage (Jiang et al., 2018a; Klein et al., 2018; Song et al., 2018). To date, diverse 2D vdW FM and AFM metal, insulators, and even semiconductors have been explored (Liu et al., 2021). FM Cr$_2$Ge$_2$Te$_6$ few-layer have demonstrated a p-type semiconducting behavior and gate-controllable magnetism (Jiang et al., 2018a; Verzhbitskiy et al., 2020); few-layer Fe$_3$GeTe$_2$ FM metal have shown high Curie temperature ($T_c$) above room temperature through ionic/protonic gating, patterning, and interfacial exchange coupling (Deng et al., 2018; Li et al., 2018; May et al., 2019; Wang et al., 2020a); magnetic topological insulator MnBi$_2$Te$_4$ has recently been reported to present layer-dependent magnetic phase and spin-flop behaviors (Deng et al., 2020; Yang et al., 2021). Alternatively, air-stable metallic 2D vdW CrTe$_2$ and CrS$_2$ with thickness-dependent magnetic orders have been directly synthesized by chemical vapor deposition (CVD) methods (Li et al., 2021; Meng et al., 2021). However, thus far, few 2D vdW magnets have been revealed luminescence and exciton behaviors, and none examining magnetic coding of photoluminescence (PL), despite the discovery of luminescence in FM CrI$_3$ and AFM NiPS$_3$ (Hwangbo et al., 2021; Kang et al., 2020; Seyler et al., 2017; Wang et al., 2021). Moreover, the PL of CrI$_3$ derives from a ligand-field-allowed d-d transition, which naturally leads to a very broad PL peak with an FWHM of 100–200 nm owing to strong vibronic coupling (Seyler et al., 2017), extremely limiting the potential for photonic devices; in addition, it is difficult to achieve magnetic/electric control of spin in AFM NiPS$_3$, due to a high critical field up to 10 T at least (Wang et al., 2021).

With the rapid development of digitalization and semiconductor optoelectronics technology, various fields have an increasing demand for on-chip lasers, so the research on high-performance and miniaturized...
on-chip lasers is of great significance (Gao et al., 2020; Kyeremateng et al., 2017). On-chip lasers have broad application prospects, such as a light source for on-chip integrated photonic chips, laser scanning, and ranging or flexible displays. Traditional directly modulated lasers (DML) have been achieved by injecting the pump current to the gain medium, and the modulation speed is affected by its volume of the active region and relaxation oscillation frequency (Dong et al., 2018; Kobayashi et al., 2013). Alternatively, topological lasers with single-mode lasing and robustness against defects have been realized by coupling of photonic gain with topological structure (Bandres et al., 2018). Additionally, atomic-thin lasers using transition metal dichalcogenides (TMDs) with strong exciton emission as optical gain medium have been recently reported (Li et al., 2017; Salehzadeh et al., 2015; Wu et al., 2015; Ye et al., 2015). However, thus far, small-volume atomic-thin lasers with intrinsic magnetism in the near-infrared (NIR) region have been still elusive, which provide numerous opportunities to realized magnetic-field controlled DML. Moreover, the NIR laser is more challenging owing to the lack of gain medium with strong NIR emission.

Here, we report the realization of magnetic control and coding of NIR whispering-gallery-mode (WGM) emission through coupling of the luminescence of 2D ferromagnetic CrI3 and SiO2 microsphere cavities. Circularly polarized WGM PL features in CrI3/microsphere are coupled with CrI3 magnetic orders. The right-handed (left-handed) polarized WGM PL is dominated in a positive (negative) magnetic field of +1 T (−1 T). Multiwavelength encoding is realized through repeatedly manipulating helicity of each WGM PL peak by magnetic field. This work opens the door to creating new atomically thin 2D magneto-optical devices for photonic integrated circuit and on-chip optical system.

RESULTS AND DISCUSSION
Coupling of magnetic orders and PL in CrI3
In 2D FM CrI3 monolayer, Cr^{3+} ions in each layer are coordinated by six nonmagnetic I^{-} ions to form an octahedron (Guo et al., 2020), which shares edges to build a honeycomb network (Figure 1A). Monolayer CrI3 is a Ising ferromagnetism arising from the Cr-I-Cr superexchange interaction, which is described by the Ising spin Hamiltonian, $H = -(1/2) \sum_{ij} J_{xy} S_{xi} S_{yi} + J_{yz} S_{zi} S_{yi} + J_{z} S_{zi} S_{x} S_{y}$, where $S_{x}, S_{y}, S_{z}$ are the spin along the x, y, z direction of Cr^{3+} at (i) sites; $J_{x}$ and $J_{z}$ are the exchange coupling term of in-plane and out-of-plane spin components and $J_{y} > J_{x} > J_{z} > 0$ in Ising FM model. In few-layer CrI3, intra-atomic d-d transitions and charge-transfer transitions owing to ligand field lead to a broad layer-dependent PL emission at ~1020 nm with an FWHM of ~100 nm (Figure S1) (Seyler et al., 2017). PL intensities show strong layer dependence and significantly increase as increasing layers (Seyler et al., 2017); therefore, thick CrI3 flakes

![Figure 1. CrI3/microsphere WGM microcavities](image)
and bulk were chose in our experiments. The helicity of the absorption and associated PL emission is correlated with the spin of the electrons at the ground states. Spin states are tied to magnetic orders of CrI$_3$ few-layers. Thus, in an out-of-plane positive magnetic field, the upper spin states are dominated and absorb right-hand polarized light, consequentially mostly giving right-hand polarized PL emission (Figure 1B); when the magnetic field turns downward, the spins of electrons are reversed, and mainly lead to left-hand polarized PL (Figure S1). But, the poor and broad PL features restrict the exploring on magnetoexciton coupling and creating photonic devices with unique functions. Optical microcavities have been widely applied to enhance light-matter interaction and suppress FWHM for realizing strong emission and lasing (Salehzadeh et al., 2015; Schwarz et al., 2014; Wu et al., 2014; Ye et al., 2015). SiO$_2$ microsphere as high-quality WGM microcavities with high Q-factor of $\sim 10^8$ can significantly enhance the coupling between gain medium and optical cavity modes (Mi et al., 2017; Zhao et al., 2018). Thus, SiO$_2$ microsphere cavities can be coupled with FM 2D layered CrI$_3$ to create narrow and strong WGM oscillation PL peaks. The coupling between circularly polarized PL emission and SiO$_2$ spherical microcavities obey the law of conservation of angular momentum; thus, the PL helicity associated with magnetic orders are preserved. In a positive magnetic field, right-hand polarization-resolved WGM oscillation PL originating from spin-up states are dominated, and vice versa (Figure 1C). Polarization-resolved Raman spectra show that thick layered CrI$_3$ bulk sample adopts a rhombohedral structure at 10 K. Raman shift of the two-fold degenerated $E_g$ mode at $\sim 107$ cm$^{-1}$ are independent of the polarization angle (Figures 2A and S2), validating the high-symmetry rhombohedral phase (Li et al., 2019; Song et al., 2019). Reflective magnetic circular dichroism (RMCD) was used to probe the long-range FM orders. A 633 nm continuous-wave laser was applied as an excitation source with the light spot size of $\sim$2 $\mu$m and 2 $\mu$W. Figures 2B and 2C show the typical RMCD curves of bare CrI$_3$ and CrI$_3$/microsphere, indicating a $T_c$ of $\sim 50$ K, and the SiO$_2$ microsphere cavities do not influence the $T_c$ of CrI$_3$.

**Magnetic control of WGMPL emission of CrI$_3$**

To obtain strong enhancements, SiO$_2$ microspheres with 10 and 12.5 $\mu$m diameter were transferred onto mechanically exfoliated thick CrI$_3$ flakes ($\sim 32$ nm) and bulk ($\sim 55$ nm) through polydimethylsiloxane
(PDMS) films (Figure S2). The Q factors of SiO2 spherical cavities were firstly estimated on MoS2/SiO2 samples. The Q factors of microsphere cavities are estimated from $Q = \lambda / \delta \lambda$, where $\delta \lambda$ is the FWHM of the WGMs, and the Q factor for the TM1,61 mode (~633 nm) can reach about 1055 (Figure S3). As shown in Figure S3B, the PL intensity increases sharply with increasing excitation light power, and the lasing behavior at the TM1,61 mode (~633 nm) is observed. Figure S3C shows PL intensity of MoS2/SiO2 and MoS2 as a function of laser power. The PL intensities of MoS2 shows a linear dependence on the excitation light power, and TM1,61 mode shows a kink indicating the onset of superlinear emission. In Figure S3D, the FWHM of TM1,61 mode decreases from 0.9 to 0.6 nm, the linewidth narrowing effect further confirms the appearance of lasing. Room temperature lasing from MoS2/sphere validates that Q-factor of SiO2 is high enough for WGM lasing (Zhao et al., 2018). WGM PL peaks in the range of ~900–1100 nm are unambiguously observed from CrI3 beneath 10 and 12.5 µm SiO2 spheres under continuous wave 633 nm excitation with 1 mW (Figure S4) and related to STAR Methods. SiO2 microsphere optical cavities enhance PL intensity of CrI3 by ~2.7 times as compared with that from the same but bare CrI3. The excitation area can be effectively reduced to enhance the coupling between the gain region and the optical mode due to the lens effect of the microspheres (Mi et al., 2017; Zhao et al., 2018), which increases the spontaneous emission efficiency and PL intensity of CrI3. In stark contrast, no oscillation peak is detected in bare CrI3 sample. Theoretical calculations demonstrate that the strong WGM PL is attributed to the first-order TM modes (TM1), as indicated by blue line (Figure S4).

To study the WGM PL in the microsphere cavity, power-dependent PL spectra were recorded on CrI3/microsphere vertical light-emitting gain structure in the range from 10 µW to 1.5 mW with a 633 nm laser excitation source at 10 K. The lensing and screening effects of the microsphere cavity increase the excitation efficiency of CrI3, the equally spaced oscillation peaks can be distinguished, even the excitation power is as low as about 10 µW (Figures S5 and S6). The WGM PL intensities linearly increase with laser power increasing. When the laser power is increased to 1.5 mW, the oscillation peak intensity increases sharply and the peak width is narrowed. The spacing between two adjacent resonant modes remains constant with increasing laser power, indicating the WGM PL originating from the same resonant modes of the sphere cavities. The PL intensities of bare CrI3 samples also linearly increase with the increase of laser power, but smaller than WGM PL intensity. The integrated PL intensities of TM1,36, TM1,37, TM1,47, and TM1,48 modes of CrI3/microsphere with 10 and 12.5 µm SiO2 spheres remain linear to the excitation power density. With further increasing laser power, the WGM oscillation peak intensity of the sample achieves maximum at 2.6 mW and drastically decrease at 3.0 mW due to laser burning of CrI3 flakes. But, no transition from spontaneous emission to amplified spontaneous emission was observed (Figures S5 and S6). This may be because the gain of CrI3 flakes is not strong enough and they are also easy to burn out under high power, resulting in no superlinear response. The PL intensity of thin CrI3 was strongly dependent on the number of layers, and decrease with decreasing thickness of CrI3. Even, no WGM peaks are observed and only PL enhancement behaviors take place if the gain of CrI3 is decreased by applying thin CrI3 flakes (Figure S7). WGM PL features are nearly same in 10 and 12.5 µm SiO2 microsphere cavities. Therefore, we focus on 10 µm SiO2 microcavities to further study the magnetic control and coding of WGM PL.

WGM PL of CrI3/microsphere can be manipulated by an out-of-plane magnetic field. Figure 3 presents the circularly polarization-resolved WGM PL features from CrI3/microsphere at 10 K, at magnetic field +1, 0, −1, and 0 T, respectively. We define the right-hand (left-hand) circularly polarized laser as RR (LL). The intensity of circularly polarized WGM PL is correlated to magnetic orders of CrI3. In a magnetic field of +1 T, the FM orders are spin-up, leading to the spin-up states are dominated; therefore, RR WGM PL is stronger than LL WGM PL (Figures 3A and 3E). As the magnetic field is lowered to 0 T, the CrI3 shows an antiferromagnetic behavior, and spin-up and spin-down states are nearly equal (Figures 3B and 3E). When magnetic field is further reversed to −1 T, spin states are also inverted, giving rise to opposite helicity and exhibiting a stronger LL component than RR (Figures 3C and 3E). When magnetic field returns back to 0 and +1 T, the WGM PL is also recovered (Figures 3D and 3A). The corresponding RMCD results clearly show the associated FM and AFM orders, which consist with the magnetic control behaviors of circularly polarization-resolved WGM PL. It should be noted that the intensity variations of right-handed and left-handed polarized PL of the CrI3/sphere are lower than the same pure CrI3 flakes (Figure 3 and Figure S1). This probably results from the decrease of magnetic anisotropy and long-range ferromagnetic orders induced by tensile strains, which are formed during the transferring SiO2 sphere onto CrI3 by a PPMA fixed-point transfer method.
Previous reports have demonstrated that tensile strains lead to an increase of bandgap associated with unchanged valence band maximum (VBM) and higher conduction band minimum (CBM) (Webster and Yan, 2018; Zhang et al., 2015). Figure S7B shows that the PL peak is blue-shifted to a high energy, which indicates that tensile strains are formed on CrI₃ flakes. Tensile strains lead to the increase of the Cr-I-Cr angle and bond length, which result in decreasing magnetic anisotropy energies and ferromagnetic orders of CrI₃. The helicity of PL emission of CrI₃ is strongly tied to magnetic orders. Therefore, the intensity variations of circularly polarized PL of CrI₃/sphere decrease.

Multiwavelength magnetic encoding

The WGM oscillation modes of sphere cavities result in a series of narrow PL at multiwavelength. We studied the multiwavelength magnetic encoding of circularly polarized PL through recycling a loop of magnetic field between +1 and -1 T. Magnetic field loop is set from +1 to 0 T and inversely increase to -1 T, and return back to +1 T (Figure 4A). The PL intensity differences between LL and RR WGM PL, defined as ΔI, show striking narrow oscillation peaks at 976, 995, 1020, 1045, and 1070 nm with FWHM between 5 and 10 nm (Figure 4B), which show an obvious magnetic-field dependent behavior. ΔI at the five distinct wavelengths are opposite at +1 and -1 T, and much larger than that at 0 T, which provide a rich platform for tri-state encoding through on/off switching of magnetic field. Figure 4B shows ten recycling of ΔI for multiwavelength magnetic encoding. The ΔI features at 1020 ± 5, 1070 ± 5, 1045 ± 5, and 995 ± 5 nm were extracted. Each wavelength presents +1, 0, and -1 tri-states at magnetic field of +1, 0, and -1T. The magnetic control behaviors of tri-states are entirely correlated to the magnetic field and display the
same shape with magnetic field setups, as shown in Figures 4C and 5. The helicity of CrI3/microsphere WGM oscillation peaks are determined by magnetic orders of CrI3 and controlled by magnetic field, which also can be used to achieve magnetic encoding function. Similar magnetic-field control behaviors have been achieved on different samples (Figures 4B and 3), although the PL intensity variations is different, which strongly validate that the SiO2 spherical cavities can generally lead to WGM peaks for CrI3 ferromagnetism and reversible magnetic control and coding of helical luminescence in the CrI3/microsphere is reproducible. The degree of polarization (DOP) from CrI3/microsphere and CrI3 is given by DOP = (I_{RR} - I_{LL})/(I_{RR} + I_{LL}), where I_{RR} and I_{LL} are the intensity of right-hand and left-hand circularly polarized PL (Li et al., 2020; Peng et al., 2017; Wang et al., 2020b). When a magnetic field of +1 T is applied, DOP of the WGM PL is around +10%, while the DOP is inversely changed to -10% in a -1 T magnetic field, owing to the reversal of the magnetization of CrI3 (Figure 6). Magnetic fields manipulate the DOP of each oscillation peak in ten recycling, clearly validating multiwavelength magnetic encoding behaviors.

**Conclusions**

In summary, we demonstrated magnetic control of NIR helical WGM PL and realized tri-state encoding at different wavelength. The lensing and screening effects of SiO2 microspheres increase the spontaneous emission efficiency of CrI3 to create unambiguous WGM oscillation peaks which is coupled with magnetic orders of CrI3. Magnetic fields manipulate and control each WGM peak, and the PL helicity and intensity difference of every WGM peak are applied to realize multiwavelength encoding through recycling magnetic fields. This work provides promising opportunities for creating magneto-optical...
devices and integrated photonic lasing with magnetic encoding function through FM 2D materials microcavity.

**Limitations of the study**
The CrI₃ sample needs to be mechanically exfoliated in a glove box, and the sample should not be exposed to the air, otherwise the sample will be hydrolyzed.

**STAR METHODS**
Detailed methods are provided in the online version of this paper and include the following:

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SUPPLEMENTAL INFORMATION

Supplemental information can be found online at https://doi.org/10.1016/j.isci.2021.103623.

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STAR METHODS

KEY RESOURCES TABLE

| REAGENT or RESOURCE | SOURCE | IDENTIFIER |
|---------------------|--------|------------|
| **Chemicals**       |        |            |
| Chromium powder     | Alfa Aesar | CAS: 7440-47-3 |
| Anhydrous iodine particles | Alfa Aesar | CAS: 7553-56-2 |
| SiO₂ microsphere    | Tianjin BigGoose Scientific Co., Ltd | Item: S2-13000 |
| **Software and algorithms** |        |            |
| Adobe Illustrator  | www.adobe.com | CC 2018 |
| Origin              | www.originlab.com | 2018 |
| **Other**           |        |            |
| Optical microscopy  | Motic  | BA310MET |
| Confocal Raman Imaging | WITec | alpha300 R |
| Thermal evaporation system | Leybold | UNIVEX250 |
| Scanning Electron Microscope | Thermo Fisher Scientific | JSM-7600F |

RESOURCE AVAILABILITY

Lead contact
Further information and requests for resources and materials should be directed to the lead contact, Prof. Bo Peng (bo_peng@uestc.edu.cn).

Materials availability
This study did not generate new unique reagents.

Date and code availability
Data reported in this paper will be shared by the lead contact upon request. No new code was generated during this study.

EXPERIMENTAL MODEL AND SUBJECT DETAILS

Sample preparation
CrI₃ and MoS₂ flakes were mechanically exfoliated from bulk crystal onto polydimethylsiloxane films and then transferred onto SiO₂/Si substrates. The commercial SiO₂ microspheres solution were dropped onto another SiO₂/Si substrates, which were heated on a heating table to make the solvent completely volatilize. Next, SiO₂ microspheres were directly transferred onto CrI₃ and MoS₂ flakes. The samples were loaded into cold head for optical measurements in glove box.

Optical measurements
The details about the optical measurements are listed under the section “quantification and statistical analysis”.

METHOD DETAILS

Materials
The SiO₂ microsphere solutions were ordered from Tianjin BigGoose Scientific Co., Ltd (Item: S2-13000). Chromium powder (99.99%) and iodine particles (99.99%) were supplied by Alfa Aesar. MoS₂ bulk crystal were bought from HQ graphene.
CrI$_3$ synthesis

About 1 g of the stoichiometric mixture of Chromium powder and iodine particles was loaded in the ampoule (16-mm inner diameter, 20-mm outer diameter, and 200-mm length), which was evacuated to a pressure of 10$^{-3}$ Pa. The ampoule was sealed and placed into a two-zone furnace with temperature gradient of 650°C to 530°C for 7 days. The CrI$_3$ crystal was obtained at the sink region of the ampoule.

Sample preparation

The details about the sample preparation are listed under the section “experimental model and subject details”.

QUANTIFICATION AND STATISTICAL ANALYSIS

Scanning electron microscopy (SEM)

The morphologies of CrI$_3$ flakes and SiO$_2$ microspheres were observed with JEOL JSM-7600M field-emission scanning electron microscope.

Raman spectroscopy

The Raman spectra were recorded by Witec Alpha 300R Plus confocal Raman microscope (1800 groves/mm grating), coupled with a closed-cycle He optical cryostat (10 K) and a 7 T magnetic field. The excitation laser of 514 nm was 2.5 mW and the integration time was 60 s. The polarization-resolved Raman spectra were obtained by rotating a half-wave plate in crossed-polarization configuration.

Photoluminescence spectroscopy

The PL signal were recorded by Witec Alpha 300R Plus confocal Raman microscope (150 groves/mm grating), coupled with a closed-cycle He optical cryostat (10 K) and a 7 T magnetic field. The excitation laser of 633 nm was 2 mW and the integration time was 15 s.

Reflective magnetic circular dichroism spectroscopy

A 633 nm HeNe laser with $\sim$2 $\mu$W was coupled to the Witec Raman system through free optical path for RMCD measurements, which was modulated by photoelastic modulator (PEM, $f_{PEM}$ = 50 KHz) and focused onto samples by a long working distance 50X objective (NA = 0.45). The reflected beams were collected by the same objective, passed through a non-polarizing beamsplitter cube into an APD photodetector.