

Spin-polarized light-emitting diodes based on CrI₃ operating without external spin injection

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Supplementary Section 1.

PL DOCP as a function of magnetic field

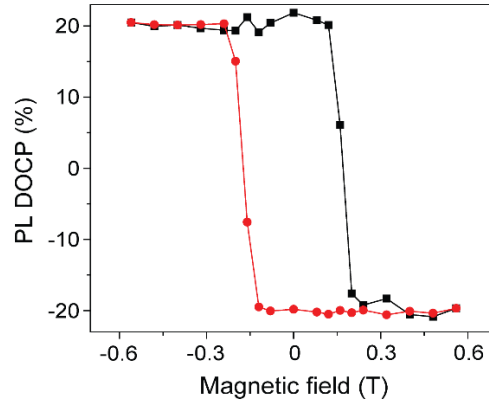


Figure S1 Change of the degree of PL polarization as a function of applied magnetic field. Red (black) data points denote the EL polarization measured when sweeping the magnetic field in the negative (positive) direction. The measurement was performed at 30 K using a linearly polarized He–Ne laser ($\lambda = 633$ nm) to excite the monolayer CrI_3 flake shown in Fig. 1b (main text).

Supplementary Section 2.

Spatially-resolved EL image of the graphene/hBN/CrI₃/hBN/graphene spin-LED

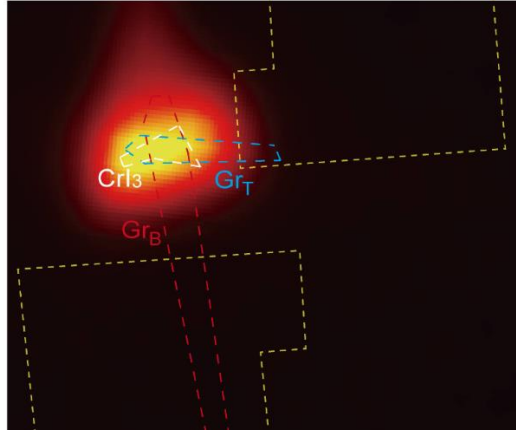


Figure S2 Spatially resolved EL image of the graphene/hBN/CrI₃/hBN/graphene spin-LED shown in Fig. 1a,b (main text), biased at 2.5 V and measured at 30 K. The top graphene (Gr_T), bottom graphene (Gr_B) and CrI₃ regions are outlined by blue, red and white dashed lines, respectively, and the EL emission is clearly localized at the graphene/CrI₃/graphene overlapping region. The image was acquired using an Andor Newton 920 CCD camera, capable of detecting light in the 300–1100 nm wavelength range.

Supplementary Section 3.

Simulated temperature distribution and current-induced temperature rise in the biased graphene/hBN/CrI₃/hBN/graphene spin-LED, obtained via finite-element analysis.

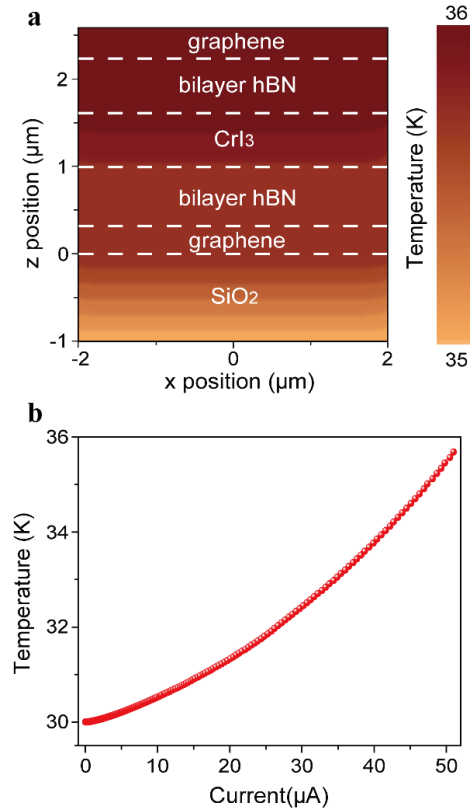


Figure S3 a. Simulated cross-sectional temperature distribution of the graphene/hBN/CrI₃/hBN/graphene spin-LED under an injected current of 50 μA . b. Simulated temperature of CrI₃ as a function of the input current injected into the graphene/hBN/CrI₃/hBN/graphene spin-LED. The power density applied to the device was calculated from the I - V characteristics shown in Fig. 3a (main text). In both panels, the LED was simulated on a cold finger at a base temperature of 30 K, and the material constants used, including thermal conductivity and specific heat, are listed in Table S1.

	Density (kg/m^3)	Specific heat ($\text{J}/\text{kg} \cdot \text{K}$)	Thermal conductivity ($\text{W}/\text{m} \cdot \text{K}$)
graphene	2267	33	287
CrI ₃	2419	92.44	1.355
hBN	2100	122.1	227

Table S1 Materials constants used in the finite element simulations¹⁻³.

Supplementary Section 4.

Temperature-dependent PL analysis of monolayer CrI_3

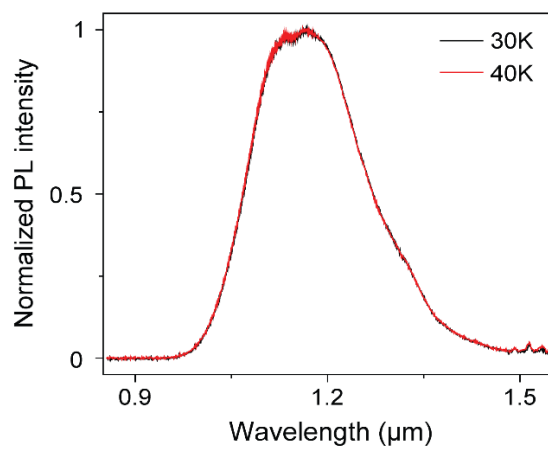


Figure S4 PL spectra of monolayer CrI_3 measured at 30 K (black) and 40 K (red). Each spectrum is normalized to its peak intensity for comparison, showing that this temperature increase induces negligible peak shift and linewidth broadening.

Supplementary Section 5.

Comparative performance of the graphene/hBN/CrI₃/hBN/graphene spin-LED and traditional III–V/II–VI spin-LEDs

	Spin injector	Emitting layer	DOCP	Temperature	Ref.
Diluted Magnetic Semiconductor	(Ga,Mn)As	InGaAs QW	8%	6K	[4]
	(Ga,Mn)As	InAs QD	1%	4.5K	[5]
	GaCrN	InGaN	6%	5K	[6]
Ferromagnetic Metal	Co _{2.4} Mn _{1.6} Ga	InGaAs QW	13%	5K	[7]
	Co/Pt	AlGaAs/GaAs	3%	20K	[8]
	CoFeB /MgO	InGaAs /GaAs QW	20-22%	25K	[9,10]
	FeCo	GaN	6%	300K	[11]
	CoFeB /MgO	InGaAs QD	19%	100K	[12]
	Not required	CrI ₃ monolayer	20%	30 K	This work

Table S2 Summary of key performance metrics and material compositions for the graphene/hBN/CrI₃/hBN/graphene spin-LED and representative III–V and II–VI spin-LEDs. The comparison includes representative material systems capable of reversing EL helicity within ± 1 T and retaining helicity after the magnetic field is removed, without requiring a large external field to maintain spin injection efficiency and suppress spin decoherence. QW, quantum well; QD, quantum dot.

Supplementary Section 6.

Stability and reproducibility of EL polarization

CrI ₃ -based spin-LEDs	EL DOCP
Device 1	20%
Device 2	18%
Device 3	17%
Device 4	21%
Device 5	23%

Table S3 Summary of EL DOCP measured from five different Gr/hBN/CrI₃/hBN/Gr heterostructures spin-LEDs. All CrI₃ flakes are monolayers, and Device 1 corresponds to the result presented in the main text. For all measurements, the spin-LEDs were first magnetized at 0.6 T and then swept back to zero field before recording the DOCP values. All measurements were performed at 30 K with an injected current of 0.5 μ A.

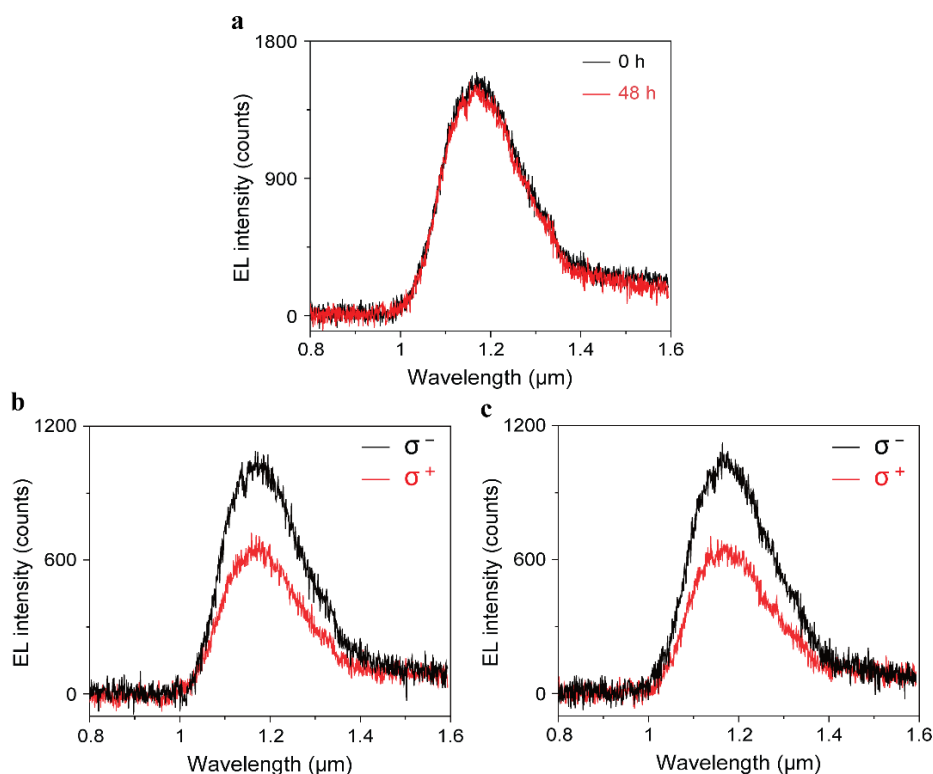


Figure S5 a. Time evolution of the EL from a graphene/hBN/CrI₃/hBN/graphene spin-LED under an injected current of 0.5 μ A. The EL intensity and lineshape remain unchanged, comparing the initial EL spectrum (black) with that measured after 48 hours of device operation (red). b-c. Polarization-resolved EL spectra for σ^- (black) and σ^+ (red) detection were recorded at a magnetic field of 0.6 T at (b) the initial measurement and (c) after 48

hours of device operation. In both cases, the EL DOCP remains around ~23%. All data were obtained from Device 5 listed in Table S3.

Supplementary Section 7.

EL DOCP hysteresis loops measured in graphene/hBN/CrI₃/hBN/graphene spin-LEDs at different bias voltages

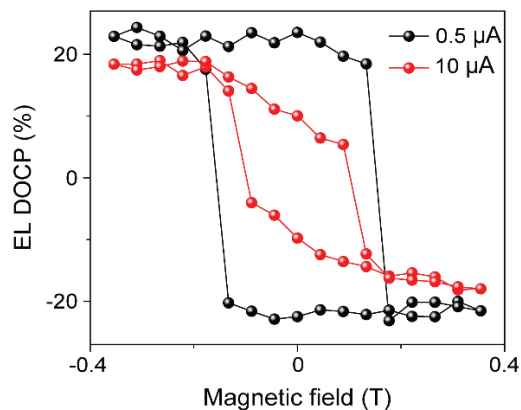


Figure S6 EL DOCP as a function of applied magnetic field for the CrI₃-based spin-LED under injected currents of 0.5 μA (black) and 10 μA (red). The results indicate that higher current injection induces a sign reversal of the EL DOCP at a lower magnetic field. All data were obtained from Device 5 listed in Table S3.

Supplementary Section 8.

Characterization of a spin-LED using bilayer CrI_3 as the emitting layer

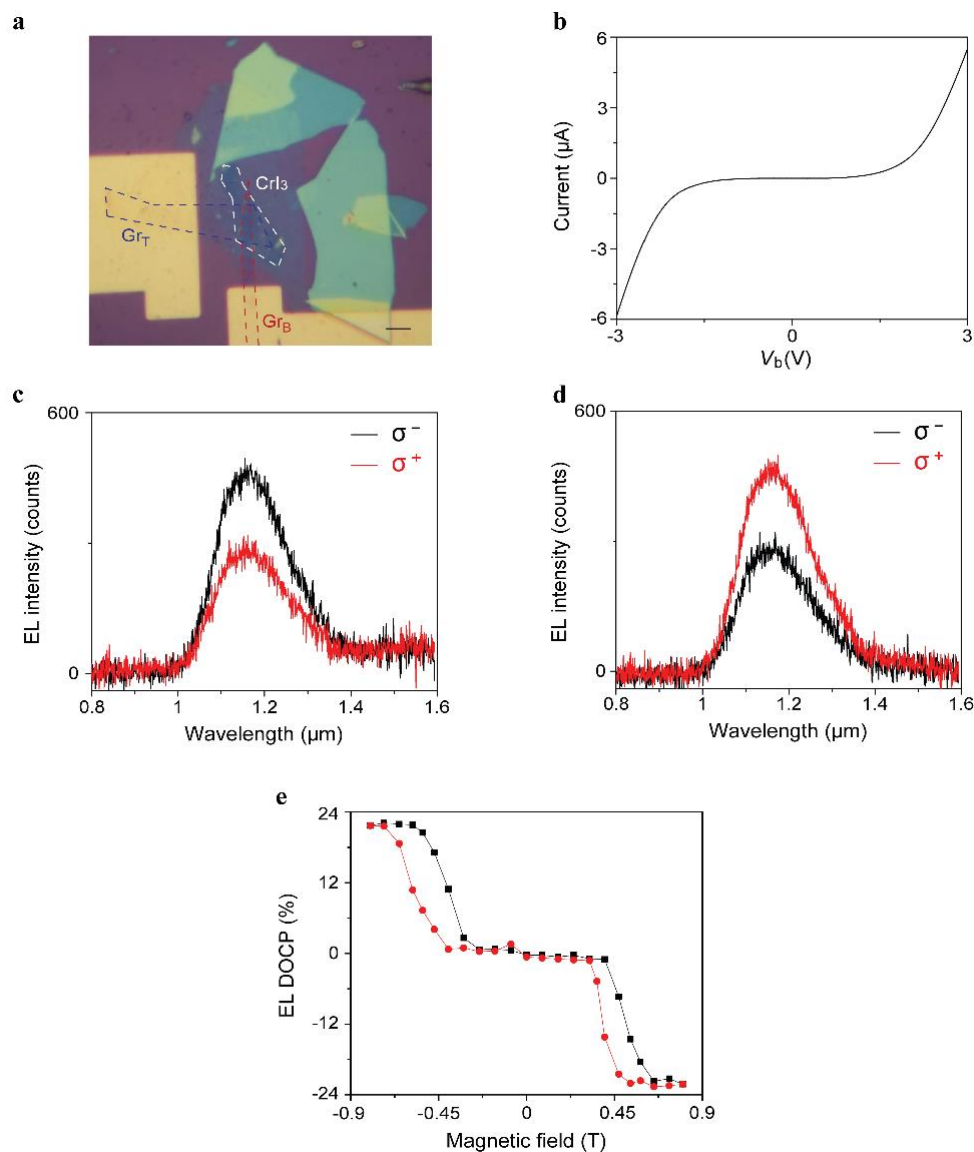


Figure S7 a. Optical microscope image of the vdW heterostructure spin-LED. The regions of top graphene (Gr_T), bottom graphene (Gr_B) and monolayer CrI_3 are defined by blue, red and white dashed lines, respectively. Scale bar, $10\ \mu\text{m}$. b. I - V_b characteristics of the light-emitting heterostructures measured under a bias applied across the graphene electrodes, with the bottom electrode grounded. c-d. Polarization-resolved EL spectra for σ^- (black) and σ^+ (red) detection, with the magnetic field at (c) $0.8\ \text{T}$ and (d) $-0.8\ \text{T}$, respectively. e. Change of the degree of EL polarization as a function of applied magnetic field. Red (black) data points denote the EL polarization measured when sweeping the magnetic field in the negative (positive) direction. Panels (c-e) were measured under an injected current of $0.5\ \mu\text{A}$.

References:

- 1 Liu, J. *et al.* Temperature dependence of thermal conductivity of giant-scale supported monolayer graphene. *Nanomaterials* **12**, 2799 (2022).
- 2 Sheng, H. H., Zhu, Y. J., Bai, D. M., Wu, X. S. & Wang, J. L. Thermoelectric properties of two-dimensional magnet CrI₃. *Nanotechnology* **31**, 315713 (2020).
- 3 Zhou, H. Q. *et al.* High thermal conductivity of suspended few-layer hexagonal boron nitride sheets. *Nano Res.* **7**, 1232-1240 (2014).
- 4 Ohno, Y. *et al.* Electrical spin injection in a ferromagnetic semiconductor heterostructure. *Nature* **402**, 790-792 (1999).
- 5 Chye, Y. *et al.* Spin injection from (Ga,Mn)As into InAs quantum dots. *Phys. Rev. B* **66**, 201301 (2002).
- 6 Banerjee, D. *et al.* Electrical spin injection using GaCrN in a GaN based spin light emitting diode. *Appl. Phys. Lett.* **103**, 242408 (2013).
- 7 Hickey, M. C. *et al.* Spin injection between epitaxial Co_{2.4}Mn_{1.6}Ga and an InGaAs quantum well. *Appl. Phys. Lett.* **86**, 252106 (2005).
- 8 Zarpellon, J. *et al.* Spin injection at remanence into III-V spin light-emitting diodes using (Co/Pt) ferromagnetic injectors. *Phys. Rev. B* **86**, 205314 (2012).
- 9 Liang, S. H. *et al.* Large and robust electrical spin injection into GaAs at zero magnetic field using an ultrathin CoFeB/MgO injector. *Phys. Rev. B* **90**, 085310 (2014).
- 10 Barate, P. *et al.* Bias dependence of the electrical spin injection into GaAs from Co-Fe-B/MgO injectors with different MgO growth processes. *Phys. Rev. Appl.* **8**, 054027 (2017).
- 11 Bhattacharya, A., Baten, M. Z., Frost, T. & Bhattacharya, P. Room temperature GaN-based edge-emitting spin-polarized light emitting diode. *IEEE Photon. Technol. Lett.* **29**, 338–341 (2017).
- 12 Giba, A. E. *et al.* Spin injection and relaxation in p-doped (In, Ga)As/GaAs quantum-dot spin light-emitting diodes at zero magnetic field. *Phys. Rev. Appl.* **14**, 034017 (2020).