

Supplementary Materials

Spin-exciton coupling modified by interfacial magnetic interactions in a van der Waals heterostructure

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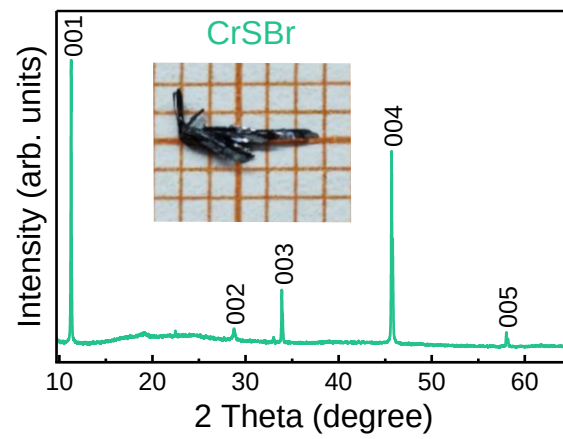


Fig. S1. Structural phase of CrSBr. XRD characterization of CrSBr single crystal. The inset shows an optical image of a typical bulk crystal.

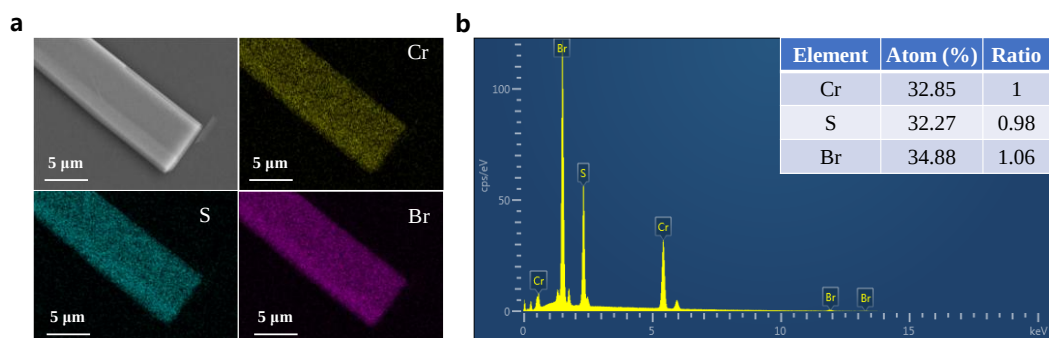


Fig. S2. Element distribution of CrSBr. **a**, SEM image and corresponding EDS elemental mappings of CrSBr. Scale bar, 5 μm . **b**, EDS spectrum of CrSBr.

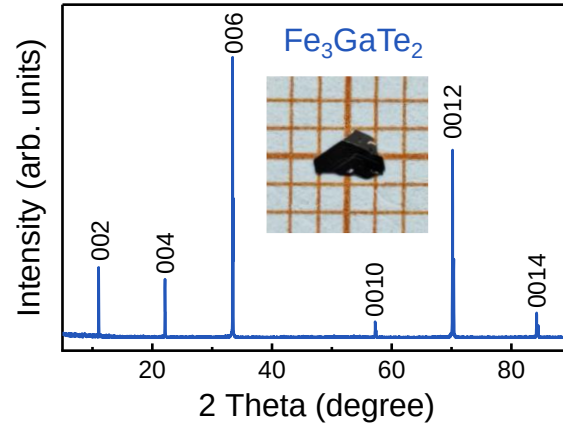


Fig. S3. Structural phase of FGT. XRD characterization of FGT single crystal. The inset shows an optical image of a typical bulk crystal.

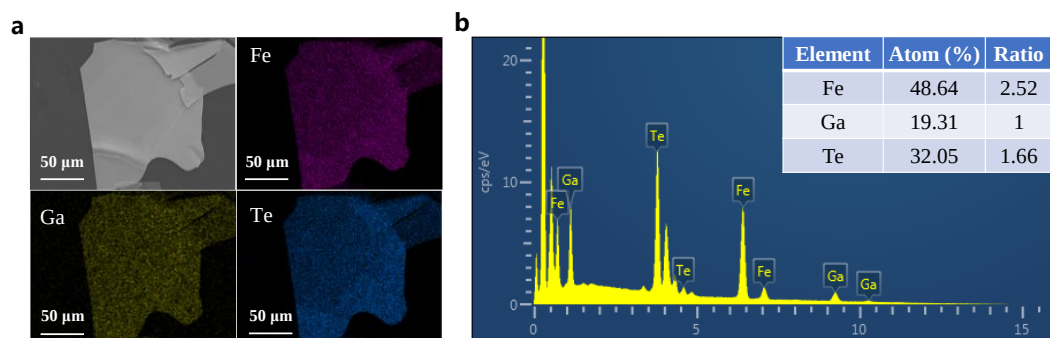


Fig. S4. Element distribution of FGT. a, SEM image and corresponding EDS elemental mappings of FGT. Scale bar, 5 μm . **b,** EDS spectrum of FGT.

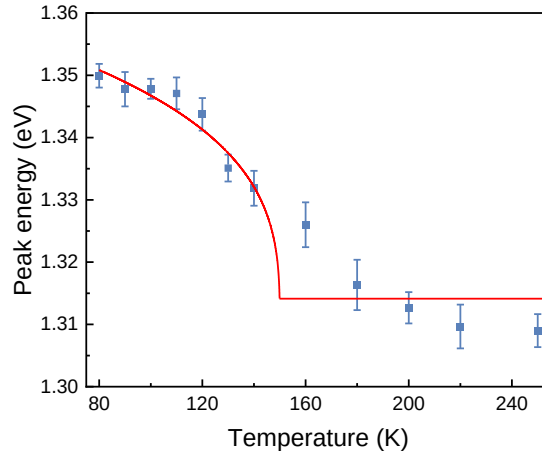


Fig. S5. Magnetic-order-dependent exciton fitting. Variations of the PL peak positions for CrSBr/FGT under different temperatures. The red solid curve is a best fit to the power law function $P^i(PL\ intensity) \propto [a + b(T - T_N)^\beta]^2$.

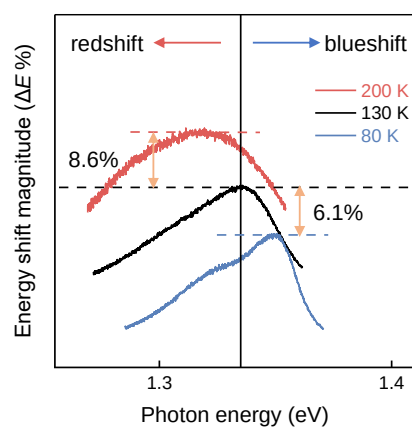


Fig. S6. Temperature-dependent modulation of exciton energy relative to the excitonic linewidth. The relative energy shift, expressed as a percentage of the excitonic linewidth (~ 100 meV), is plotted for three representative temperatures.

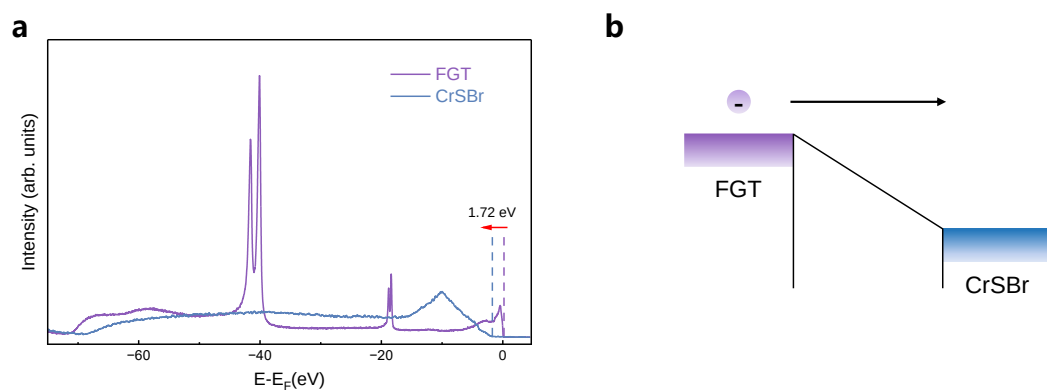


Fig. S7. Band information for heterostructure. a, UPS spectra of FGT and CrSBr. **b**, Charge transfer diagram.

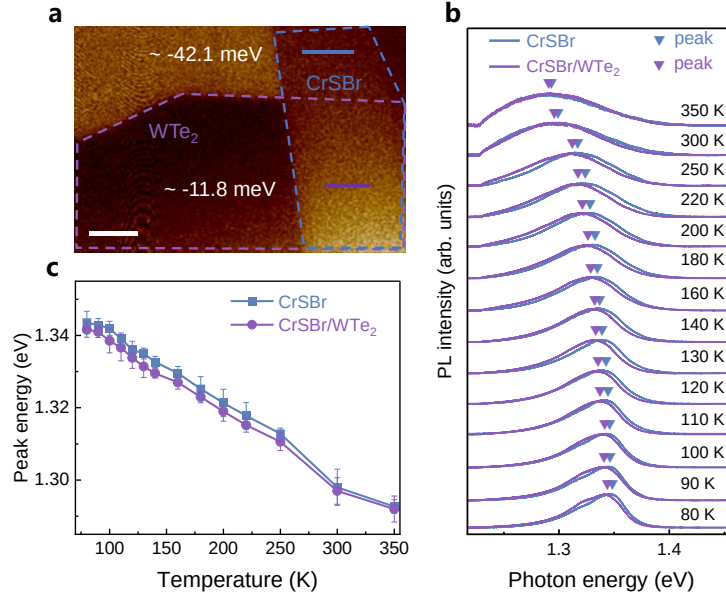


Fig. S8. KPFM and temperature-dependent PL for CrSBr/WTe₂. **a**, KPFM topography and work function plots of CrSBr/WTe₂ heterostructures. The blue and purple dashed lines mark the areas of CrSBr and WTe₂, respectively. Scale bar, 5 μ m. **b**, Temperature-dependent PL spectra for pristine CrSBr and CrSBr/WTe₂. The triangles correspond to the energy position of their PL peaks. **c**, Variations of the PL peak positions for pristine CrSBr and CrSBr/WTe₂ under different temperatures. Error bars represent the standard deviation of three measurements.

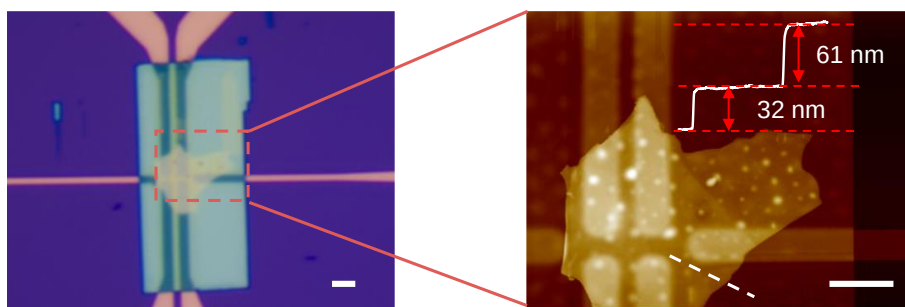


Fig. S9. Device structure. Optical image and atomic force microscopy of FGT/CrSBr heterostructure. Scale bar, 5 μm .

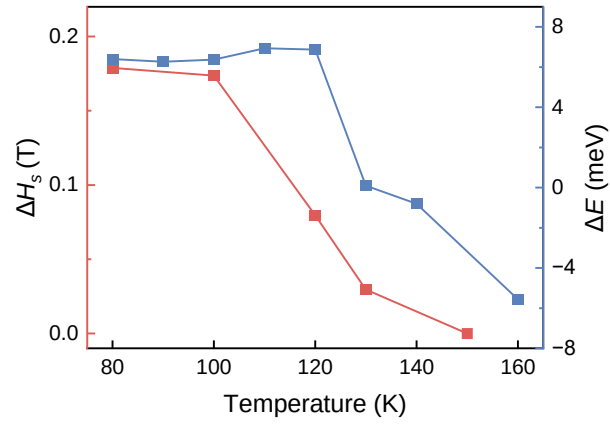


Fig. S10. Correlation between spin-flip and exciton energy. Temperature-dependent evolution relationship between the extracted spin-flip field and the exciton energy difference.

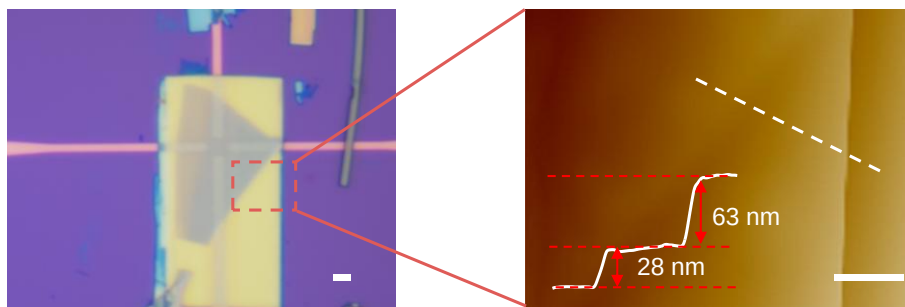


Fig. S11. Device structure. Optical image and atomic force microscopy of CrSBr/FGT heterostructure. Scale bar, 5 μm .

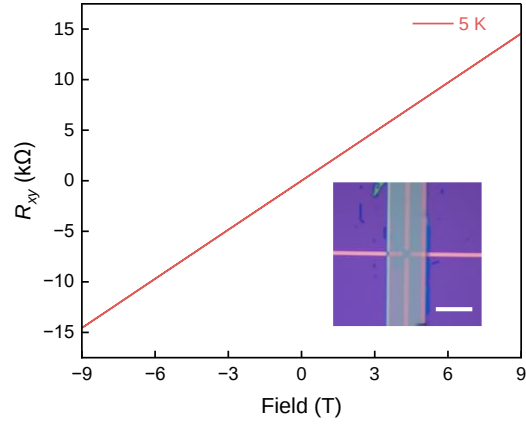


Fig. S12. Hall measurement of CrSBr. R_{xy} of the pristine CrSBr versus magnetic field at 5 K, measured with a perpendicular applied magnetic field. The inset shows an optical image of the CrSBr device. Scale bar, 20 μm .

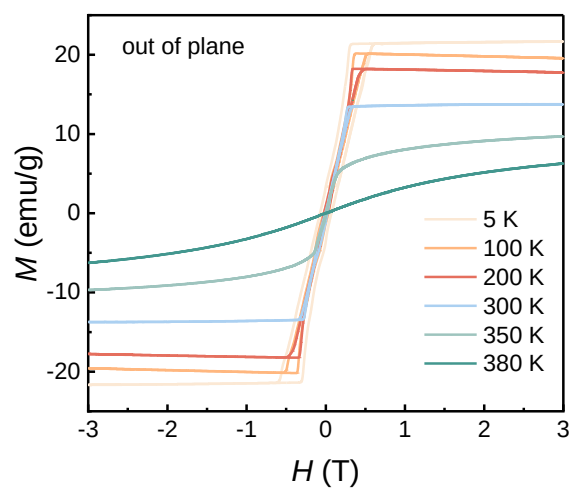


Fig. S13. Out-of-plane M - H for FGT. Field-dependent magnetization plot of FGT at varying temperatures, under a magnetic field along the out-of-plane of the crystal.

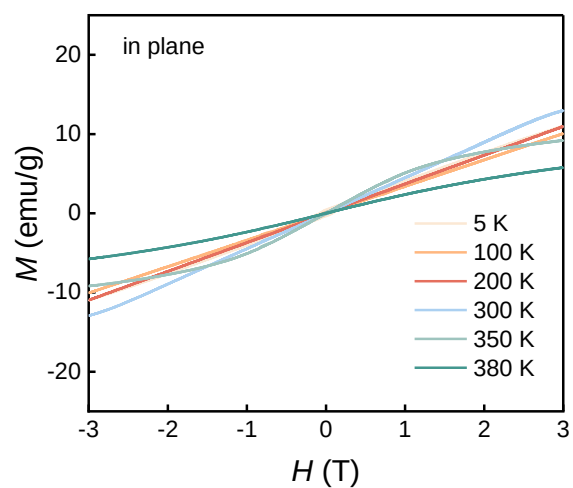


Fig. S14. In-plane M - H for FGT. Field-dependent magnetization plot of FGT at varying temperatures, under a magnetic field along the in-plane of the crystal.

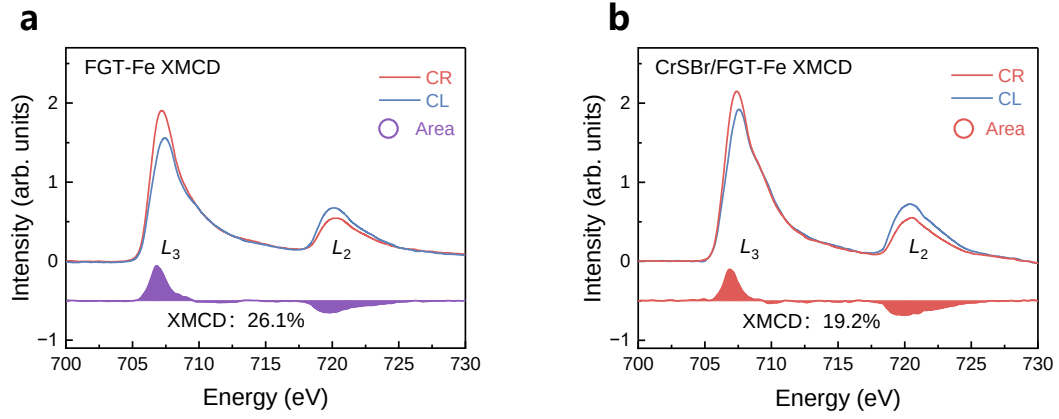


Fig. S15. XMCD signal. XMCD of Fe- L edge in FGT (a) and in CrSBr/FGT (b) extracted from the dark areas.

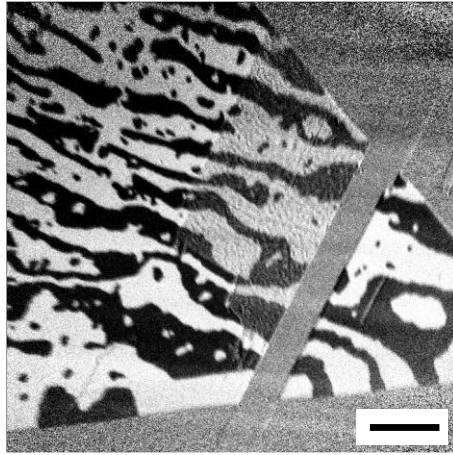


Fig. S16. XMCD-PEEM. Fe-*L* edge XMCD-PEEM image at 300 K. Scale bar, 4 μm .

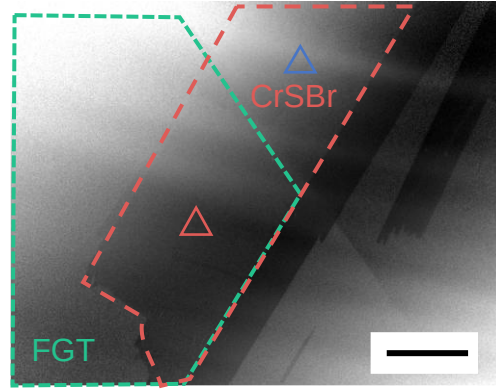


Fig. S17. XMLD-PEEM. Cr-*L* edge XMLD-PEEM image acquired with a 16° X-ray incidence angle relative to the sample surface. Scale bar, 4 μm . The triangle frames marked by blue and red respectively represent the pristine and heterostructure areas used for XMLD data extraction.

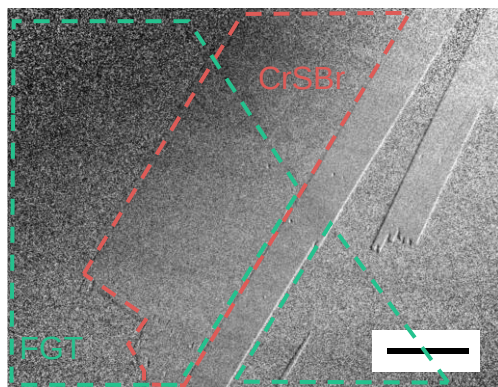


Fig. S18. XMCD-PEEM. Cr-*L* edge XMCD-PEEM image at 100 K. The red and green dashed lines mark the areas of CrSBr and FGT, respectively. Scale bar, 4 μm .

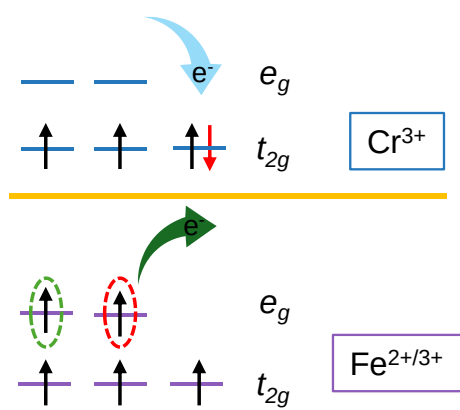


Fig. S19. Schematic diagram of charge transfer. Illustration of orbital-mediated charge transfer mechanism.

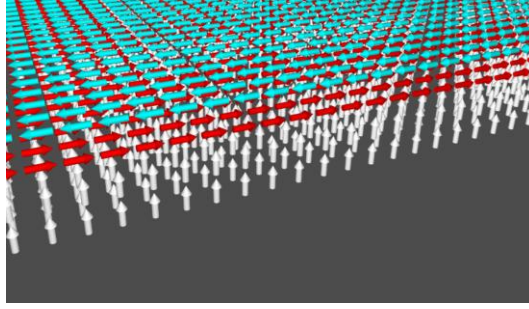


Fig. S20. Micromagnetic simulations. Schematic of the four-layer model, consisting of two out-of-plane ferromagnetic FGT layers and two in-plane antiferromagnetic FGT layers.

Table S1. Amount of electron transfer. DFT calculated electronic transfer in CrSBr/FGT heterostructure. The symbol “+” denotes electron gain, while “−” represents electron loss. Unit: $e/\text{\AA}^2$.

CrSBr	FGT
+0.007	−0.007