Supplementary Materials for

Large Room-Temperature Magnetoresistance in van der Waals Ferromagnet/Semiconductor Junctions

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Supplementary Note 1: Methods

Materials and device fabrication. The high-quality vdW bulk single-crystal WSe_2 and hBN were purchased from HQ Graphene, while Fe_3GaTe_2 was grown by the self-flux methods. To fabricate a $Fe_3GaTe_2/WSe_2/Fe_3GaTe_2$ MTJ, a Fe_3GaTe_2 flake was firstly exfoliated onto polydimethylsiloxane (PDMS) stamps by adhesive tape. Under an optical microscope, the Fe_3GaTe_2 flake with appropriate thickness and shape was chosen for transfer onto a 300 nm thick SiO_2/Si by using a position-controllable dry transfer method. Then, using the same method, a WSe_2 flake was transferred onto the Fe_3GaTe_2 flake, followed by another thicker Fe_3GaTe_2 flake to fabricate a vdW heterojunction. To prevent the Fe_3GaTe_2 from oxidation, a hBN layer was used to cap the whole heterostructure. Finally, the device was annealed at 120 ℃ for 10 minutes to reduce the bubbles between the layers and ensure close contact between the layers. Notably, the whole transfer processes were performed in a nitrogen-filled glovebox with a concentration of less than 0.1 ppm of oxygen and water to ensure a clean interface. The source and drain electrode regions were pre-patterned by standard photolithography, and Cr/Au (10/40 nm) layers were deposited using an ultrahigh vacuum magnetron sputtering system, followed by a lift-off process. By a similar process, a Fe_3GaTe_2/hBN heterostructure was stamped onto four pre-patterned Cr/Au (10/15 nm) electrodes on a 300 nm thick SiO_2/Si substrate to form a Hall-bar device. The electrodes of the Hall-bar device were patterned by standard electron beam lithography (EBL) and an ultrahigh vacuum magnetron sputtering, followed by a lift-off process.

Electrical, optical and microscopy measurements. The electrical measurements were carried out in a Model CRX-VF Cryogenic Probe Station (Lake Shore Cryotronics, Inc.) with a ±2.25 T out-of-plane perpendicular magnetic field. The instrument operation temperature varies from 10 to 400 K. The AHE was measured by the combination of Keithley model 2602B sourcemeter and Keithley model 2182 A nanovoltmeter. The electrical transport properties of MTJs were measured by a semiconductor characterization system (Agilent Technology B1500A). The thickness of Fe_3GaTe_2 and WSe_2 flakes were determined by an AFM (Bruker Multimode 8). The $Fe_3GaTe_2/WSe_2/Fe_3GaTe_2$ device optical image was obtained using an Olympus optical microscope. Raman and photoluminescence spectra of WSe_2 and Fe_3GaTe_2 flakes were obtained by optical microscopy (Renishaw inVia-Reflex) with excitation by a 532 nm laser.

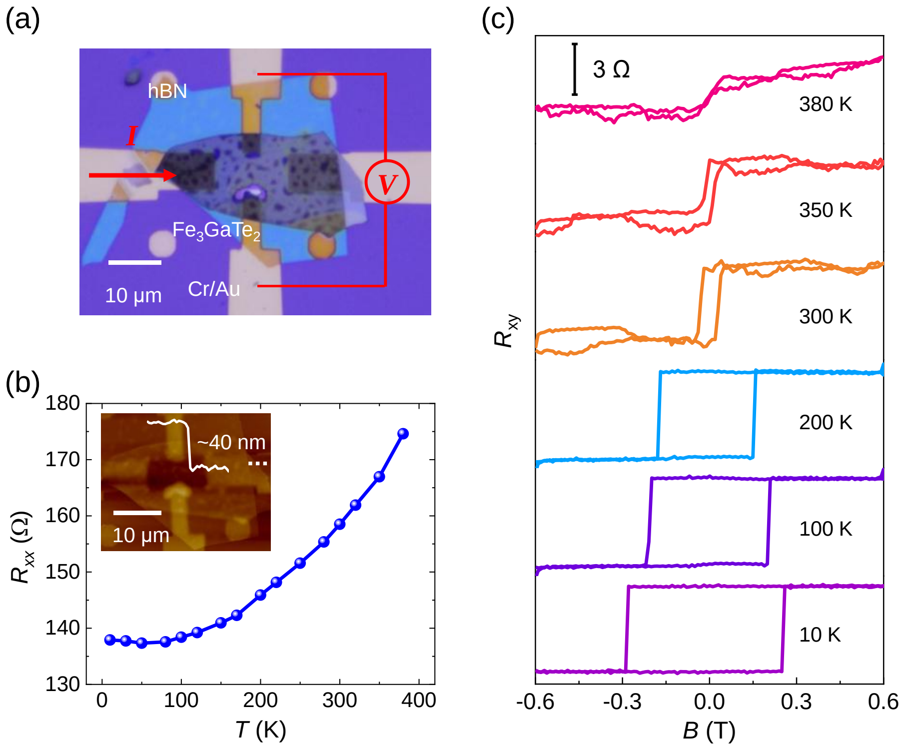

Figure S1: (a) The optical image of a thick (40 nm) Fe_3GaTe_2 Hall-bar device capped by hBN. Scale bar: 10 μm. The direction of the current and the connection of the voltmeter are shown in red. (b) Longitudinal resistance R_{xx} versus temperature from 10 to 380 K. Upper-left inset: AFM image and height profile of the Fe_3GaTe_2 Hall-bar device. Scale bar: 10 μm. (c) Hall resistance R_{xy} versus perpendicular magnetic field (B) at different temperatures ranging from 10 to 380 K. The bias current I_{xx} is fixed at 10 μA.

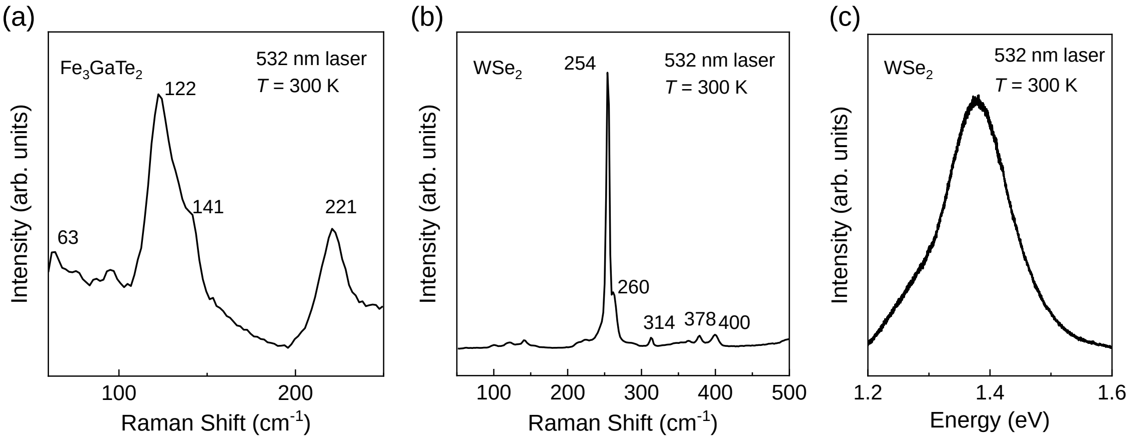

Figure S2: (a) Raman spectrum of a bulk Fe_3GaTe_2 flake (T = 300 K, λ = 532 nm). (b) Raman spectrum of a bulk WSe_2 flake (T = 300 K, λ = 532 nm). (c) Photoluminescence (PL) spectrum of a bulk WSe_2 flake (T = 300 K, λ = 532 nm). The spectrum shows the band edge emission of WSe_2, which is centred at about 1.38 eV.

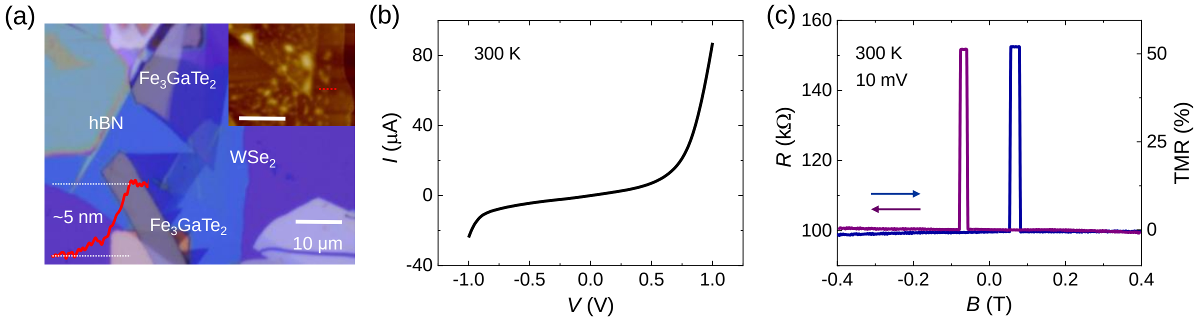

Figure S3: (a) Optical image and AFM image (upper right inset) of another $Fe_3GaTe_2/WSe_2/Fe_3GaTe_2$ MTJ device with a thin (5 nm) spacer layer. Scale bar: 10 μm. Lower-left inset: the height profile of the spacer layer WSe_2. (b) *I*-*V* characteristics of the device at T = 300 K. (c) Room temperature R and TMR versus B at 10 mV bias.

Supplementary Note 2: Temperature-dependent TMR of another device

Figures S4a-b show the TMR curves of another device measured at various temperatures. An extremely large TMR of 210% in the device is observed at T = 10 K. Same as the device in the main text, the magnitude of the TMR decreases and vanishes above T_C with increasing temperature (Fig. S4c). Interestingly, the TMR of this device (210%, 10K) is larger than that of the device in the main text (164%, 10K) at a low temperature, but the TMR of this device (53%, 300K) is smaller than that of the device in the main text (85%, 300K) at room temperature, which indicates that the TMR of this device decays more significantly with increasing temperature. This is caused by the difference in T_C of Fe_3GeTe_2 between the two devices. Fitted by power-law $P = P_0(1\text{-}T/T_C)^{\beta}$ (Fig. S4d), we get a T_C = 357 K of Fe_3GeTe_2 in this device, which is lower than that of the device in the main text (T_C = 400 K). From the literature [S1], we know that the T_C of Fe_3GeTe_2 decreases monotonically with decreasing thickness. Thinner Fe_3GeTe_2 layers (~8nm bottom layer and ~9nm top layer) in this device lead to a lower T_C, resulting in a smaller TMR at room temperature.

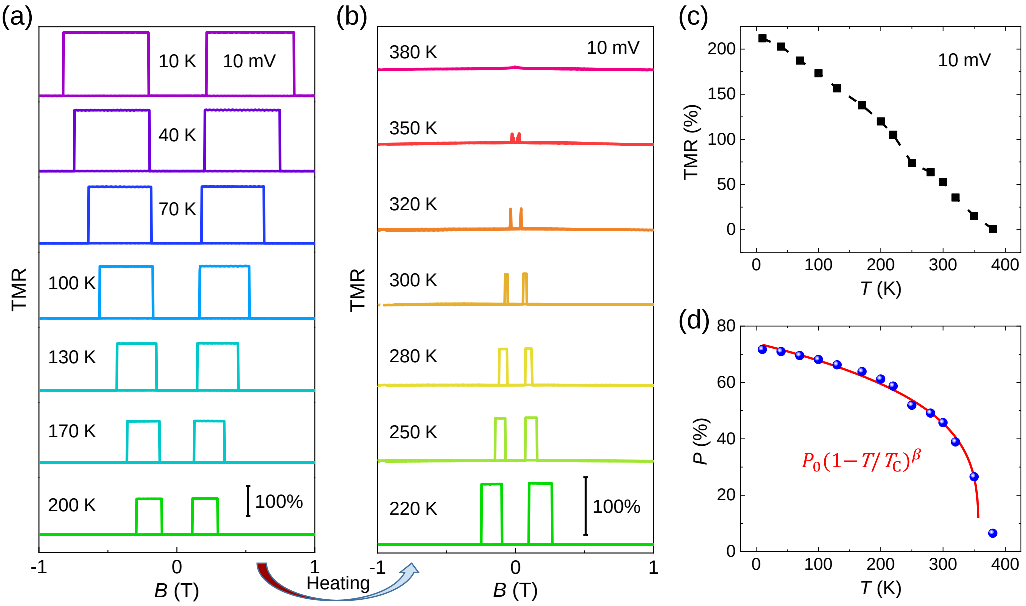

Figure S4: (a-b) TMR versus B of another device at different temperatures ranging from 10 to 380 K. (c) TMR versus temperature of the device at V = 10 mV. The values of TMR are obtained from parts (a-b). (d) Spin polarization (P) versus temperature of the device. The fitting curve follows the form of $P = P_0(1\text{-}T/T_C)^{\beta}$.

Reference

[S1]Zhang G, Guo F, Wu H, Wen X, Yang L, Jin W, Zhang W and Chang H 2022 *Nat. Commun.* **13** 5067