

Angular-independent magnetoresistance in Fe_3GeTe_2 -based van der Waals spintronic heterostructures

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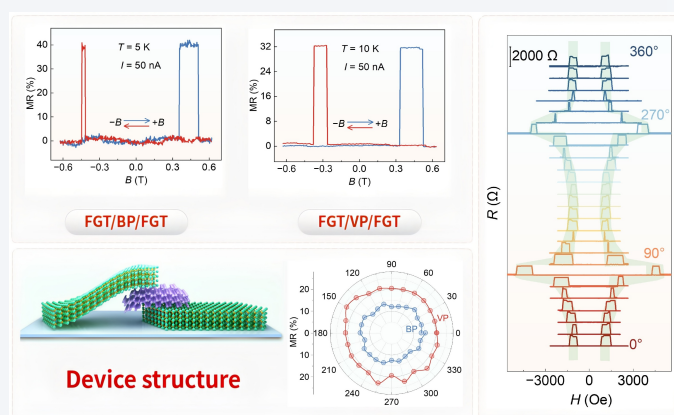
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 **Cite this article:** *Nano Research*, 2026, 19, 94908859. <https://doi.org/10.26599/NR.2026.94908859>

ABSTRACT: Two-dimensional (2D) spintronic devices based on van der Waals (vdW) heterostructures have potential applications in magnetic sensing and data storage. Here, we report vdW heterostructures fabricated by stacking an exfoliated ultrathin black phosphorus (BP) or violet phosphorus (VP) spacer between two exfoliated ferromagnetic Fe_3GeTe_2 (FGT) electrodes. The devices achieve high magnetoresistance (MR) ratios of 40.4% and 31.5% for the FGT/BP/FGT and FGT/VP/FGT heterostructures at low temperature, respectively. The magnitude of MR can be tuned by bias current and spacer thickness. Moreover, magnetotransport measurements confirm robust spin valve effect, showing angular-independent MR in FGT-based heterostructures. Our work opens a new avenue for the application of monoelemental phosphorus as spacer in novel spintronic devices.

KEYWORDS: magnetoresistance (MR), angular-independent, Fe_3GeTe_2 , spin valve, phosphorus



1 Introduction

Spin valves are essential spintronic components, widely utilized in applications such as memory devices [1, 2], logic gates [3], and magnetic sensors [4]. Their fundamental operation relies on the magnetoresistance (MR) effect, whereby the device resistance changes according to the relative alignment—parallel or antiparallel—of two ferromagnetic electrodes [5–8]. Achieving a substantial MR ratio requires maintaining high spin polarization of electrons across the spacer and interfaces, which remains a central challenge in spintronics [9, 10].

All-two-dimensional (2D) spin valves based on van der Waals

(vdW) heterostructures not only ensure high-quality interfaces assembled by pristine 2D crystals without degradation, but also avoid the common obstacles in traditional magnetic heterostructure growth such as lattice mismatch and atom interdiffusion [11, 12]. In particular, Fe_3GeTe_2 (FGT) exhibit superior characteristics among the identified vdW magnetic materials, including metallic properties [13, 14], perpendicular magnetic anisotropy [15–19], enhanced stability at atomic scales [20, 21], and a relatively high Curie temperature (T_C) of 220 K [22–25]. These features make FGT a promising ferromagnetic electrode for vdW spin-valve and magnetic-tunnel-junction-type heterostructures [26]. Nevertheless, the preservation of its magnetic robustness in vertical spin-dependent transport must be verified at the device level. Angular-independent MR is therefore important because it demonstrates that the resistance signal remains governed by two bistable magnetization configurations rather than by continuous magnetization rotation under tilted magnetic fields, enabling stable magnetic-state readout for nonvolatile spintronic applications [27]. Previous studies have shown that methods to achieve a larger MR ratio in 2D spin valves include increasing the spin polarizability of

Received: March 2, 2026; Revised: April 29, 2026

Accepted: May 21, 2026

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2D magnetic materials and selecting more suitable spacers (such as longer spin diffusion lengths, λ_s). In the research process of spin valves based on FGT, researchers selected multiple types of 2D semiconductors, such as MoS_2 , WS_2 and WSe_2 , as spin transport channel spacers, which showed MR values ranging from 0.45% to 25% at 10 K [28–30]. These values remain far below the thresholds necessary for industrial applications. Advancing spin-valve performance therefore depends critically on the identification of superior spacer materials or device architectures.

Monoelemental vdW crystals such as black phosphorus (BP) and violet phosphorus (VP) provide a highly promising platform for spin valve spacers. Compared with graphene (a zero-bandgap semimetal) and *h*-BN (a ~ 6 eV wide-bandgap insulator) [31], black phosphorus is a single-element layered material bonded by weak vdW forces between layers, allowing easy exfoliation to obtain two-dimensional black phosphorus with single or few layers. Bulk black phosphorus possesses a direct bandgap (~ 0.3 eV), and the bandgap gradually increases with decreasing layer numbers, indicating that BP has a tunable bandgap (2 eV in the monolayer) [32]. Furthermore, BP has high carrier mobility, weak spin-orbit coupling and long spin diffusion lengths (~ 6.15 μm [33]). It has been theoretically predicted that a relatively large MR ratio in BP-based spin valves, while a MR ratio of 0.57% was experimentally observed in NiFe/BP/NiFe spin valve at low temperature of 4 K [34]. Recently, M. Galbiati et al. experimentally observed a high MR ratio (500%) in the Co/BP/Co spin valve at 2 K [35]. As a layered vdW semiconductor, VP shows superior thermal stability compared with BP. Also, monolayer VP is expected to have a wide bandgap (~ 2.5 eV) and high carrier mobility (holes up to ~ 7000 $\text{cm}^2/(\text{V}\cdot\text{s})$) [36]. These properties position BP and VP as highly promising spacer candidates for next-generation spintronic devices.

We systematically investigated the magnetotransport properties of the FGT/BP(VP)/FGT vdW heterostructure. These heterostructures exhibit a notable MR ratio exceeding 40% at 5 K, successfully demonstrating a vertical spin-valve structure based on BP and VP and yielding a relatively large MR response. Moreover, by rotating the magnetic field direction, we discovered isotropic MR behavior—an important finding that originates from strong PMA and domain-wall-motion-mediated magnetization reversal of FGT. These results confirm the viability of BP and VP as efficient spin-transport channels and underscore their potential for application in spin valves with angular-independent MR characteristics.

2 Experimental section

2.1 Sample preparation and characterization

High-quality FGT single crystals were synthesized using chemical vapor transport (CVT) method with iodine as the transport agent. A mixture of high-purity elemental powders—Fe (99.9%), Ge (99.999%), and Te (99.999%)—were combined with I_2 in a molar ratio of 3:1:2:0.1, sealed in a quartz tube, and placed in a two-zone tube furnace. The growth process involved a carefully controlled heating sequence. The entire furnace was initially heated to 625 °C at a rate of 1 °C/min to minimize thermal stress. Then, the source zone was rapidly heated to 800 °C within 1 h, establishing a stable temperature gradient of 800 (source) to 625 °C (growth zone). The temperature profile was maintained for 4 days to facilitate vapor transport and single crystal nucleation. BP, VP, and *h*-BN crystals

are commercially obtained from Nanjing Muke Nano Technology Co., Ltd.

2.2 Device preparation

Devices were fabricated on a SiO_2/Si substrate with pre-patterned Ti/Au (5/15 nm) electrodes, defined by standard lithography and thermal evaporation. Single-crystal FGT was mechanically exfoliated into few-layer flakes using adhesive tape. A flake of suitable thickness and morphology was selected via optical microscopy while adhered to a polydimethylsiloxane (PDMS) stamp and subsequently transferred onto the electrodes using a dry transfer method. The vdW heterostructure was fabricated by sequentially transferring a spacer (either BP or VP) and the top FGT layer onto the bottom FGT layer. To protect the structure from oxidation, it was encapsulated with an *h*-BN layer, followed by baking at 120 °C for 10 min to improve the interfacial contact. All exfoliation and transfer steps were carried out in an Ar-filled glove box ($\text{O}_2 < 0.1$ ppm, $\text{H}_2\text{O} < 0.1$ ppm).

2.3 Device measurements

Electrical and magneto-transport measurements were carried out using a physical property measurement system (ColdTUBE™, MultiFields). For the DC transport characterization, a two-terminal configuration was employed utilizing a precision current source (Keithley 6221) and a nanovoltmeter (Keithley 2182A).

3 Results and discussion

In this study, we have fabricated FGT single crystals via CVT method. The crystal structure of FGT was determined by X-ray diffraction (XRD) analysis. There are only (00l) diffraction peaks observed in XRD pattern (Fig. S1(a) in the Electronic Supplementary Material (ESM)), demonstrating the oriented growth of FGT single crystal along the *c*-axis. The sharp diffraction peaks further verify the high crystallinity of the FGT single crystal. In addition, the Raman spectra exhibit three characteristic peaks of FGT located at approximately 95.8, 124.0, and 138.0 cm^{-1} , corresponding respectively to E_{2g}^1 , E_{2g}^2 , and A_{1g}^1 modes [37], as shown in Fig. S1(b) in the ESM. Figure 1(a) shows a cross-sectional high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) image of mechanically exfoliated FGT nanoflakes, showing the periodically ordered atomic arrangement. In the atomic lattice structure of Fe_3GeTe_2 , Fe_3Ge slabs are sandwiched between Te atomic layers, forming a classic sandwich structure, consistent with the crystal structure. The measured interplanar spacings corresponding to the (002) and (110) planes are approximately 0.81 and 0.35 nm. The above results reveal that high-quality FGT single crystals have been successfully synthesized via CVT. The anomalous Hall effect (AHE) is a key tool for probing the magnetic transport properties of devices. We prepared three FGT nanoflake devices with different thicknesses (11, 20, and 36 nm), as shown in Fig. S2 in the ESM. Figures 1(b)–1(d) present anomalous Hall resistance (R_{xy}) as a function of temperature for three devices, measured under an out-of-plane (OOP) magnetic field sweeping between ± 0.5 T. In the temperature range of 10 to 200 K, R_{xy} –*H* curves remain approximately square-shape, demonstrating strong PMA in FGT nanoflakes. Furthermore, three FGT nanoflakes show similar temperature-dependence of the coercivity H_C (Fig. 1(e)). Due to the thermal fluctuation, H_C decreases with increasing the temperature and vanishes at about 200 K, suggesting $T_C \approx 200$ K.

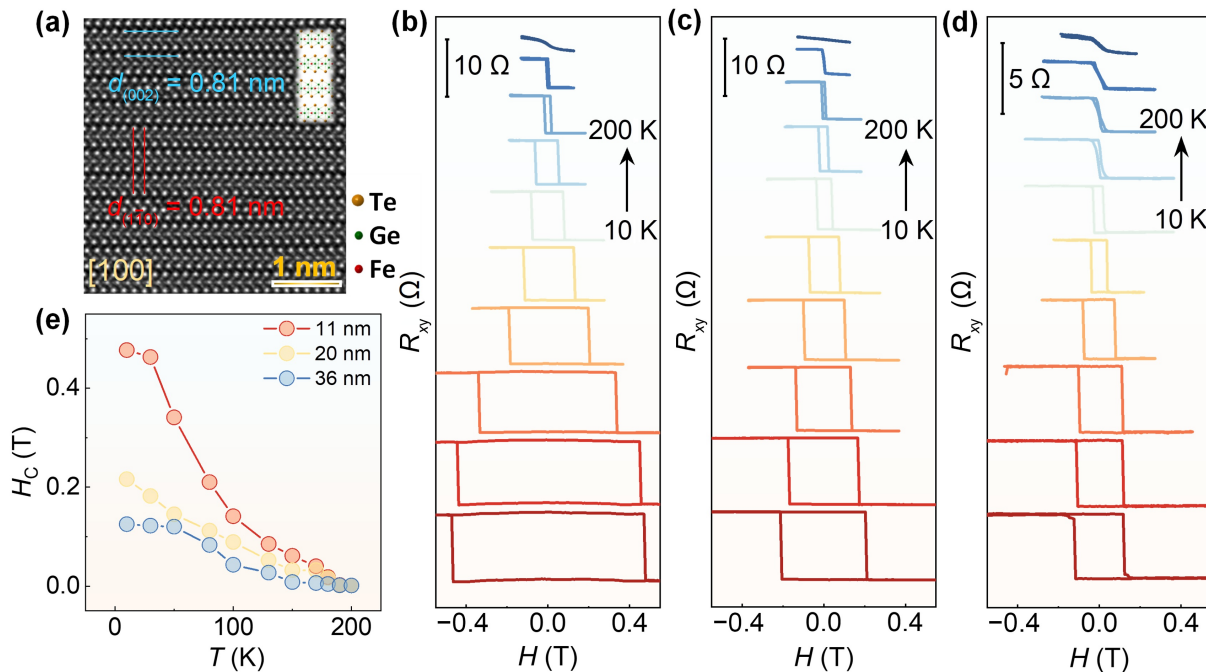


Figure 1 Crystal structure and magnetotransport properties of typical exfoliated FGT nanoflakes. (a) HAADF-STEM image (left view) and the atomic lattice structure (right view) of FGT. (b)–(d) Hall resistance R_{xy} versus OOP magnetic field for the FGT Hall bar devices with three different thicknesses at the temperature ranging from 10 to 200 K. (e) H_c as a function of temperature.

The H_c in the 11 nm FGT nanoflake reaches ~ 4950 Oe at 10 K, while exhibiting a pronounced negative correlation with sample thickness—a trend attributed primarily to surface spin disorder pinning and interlayer exchange coupling. The magnetic properties were further verified via magneto-optical Kerr effect (MOKE) measurements (Fig. S3 in the ESM). The relatively high T_c , large H_c , and robust PMA of exfoliable FGT nanoflakes make them a viable candidate for high-performance ferromagnetic electrodes.

Subsequently, FGT was employed in spintronic heterostructures to investigate their magnetotransport properties. Figures 2(a) and 2(d) show optical images and schematic diagrams of FGT/BP(VP)/FGT heterostructure, respectively. Atomic force microscopy (AFM) characterization of the heterostructures (Fig. S4 in the ESM) determined the thicknesses of the nonmagnetic spacers, with BP and VP measured at 7 and 8.5 nm, respectively. Raman spectroscopy further confirms the phase identity of the spacers (Fig. S5 in the ESM). The characteristic peaks of BP appear at 361, 438, and 466 cm^{-1} corresponding to A_g^1 , B_{2g} , and A_g^2 modes [38], respectively, whereas VP exhibits vibrational features consistent with those reported for violet phosphorus [39]. Figures 2(b) and 2(e) present nonlinear I - V curves measured at 10 K, indicating that no Ohmic contact between FGT and BP (VP). As shown in the inset of Figs. 2(b) and 2(e), the resistance presents a non-monotonic temperature dependence: it initially decreases at low temperature and then increases at higher temperature. Such an increasing trend may be related to the strong hybridization between P atoms and Fe/Ge/Te atoms [40]. Next, we measured the magnetotransport property of heterostructures with OOP magnetic field at low temperature under a bias current of 50 nA, as shown in Figs. 2(c) and 2(f). At the positive saturated magnetic field, parallel (P) alignment of the magnetic moments in the upper and lower FGT give rise to a low resistance (R_p). With decreasing magnetic field, there appears a plateau on MR curves because of the difference of H_c in the upper and lower FGT, while antiparallel

(AP) alignment of the magnetic moments on the plateau lead to a high resistance (R_{AP}) [41–43]. Based on MR curves, the MR ratio can be calculated by Eq. (1)

$$\text{MR} = \frac{R_{AP} - R_p}{R_p} \quad (1)$$

The MR ratio for FGT/BP/FGT and FGT/VP/FGT heterostructure are 40.4% and 31.5%, which are comparable to those reported in FGT-based devices [28–30]. Furthermore, the coercivities of FGT are different with decreasing and increasing magnetic field. It has been reported in the literature that the magnetization reversal is dominated by domain wall motion in Fe_3GeTe_2 [44].

In FGT/BP/FGT and FGT/VP/FGT heterostructure, the observation of asymmetric magnetic switching behavior may be related with the pinning effect arising from defects and impurities. Notably, increasing the spacer thickness drastically reduces MR ratio. For instance, the MR ratio of FGT/BP (10 nm)/FGT decreases by two orders of magnitude compared to the heterostructure with a thinner spacer of 7 nm (Figs. S6 and S7 in the ESM). As the BP spacer thickness increases, longer transport path of spin-polarized electrons induces enhanced spin scattering in the spacer, which degrades the spin polarization and thus significantly lowers the MR ratio, highlighting the critical role of spacer dimensions in magnetotransport behavior.

The bias currents dependence of fabrication FGT/BP/FGT and FGT/VP/FGT heterostructures were measured by MR versus OOP field curves. As shown in Fig. 3(a), the pronounced spin valve effect is observed across bias currents from 50 nA to 10 μA at 5 K in the heterostructure with BP spacer. Furthermore, FGT/VP/FGT heterostructure shows the spin valve effect in a wide bias currents range from 1 nA to 5 μA at 10 K. Notably, the effect remains stable even at very low bias currents down to 1 nA, indicating the potential applications in the lower-power device. At a bias current

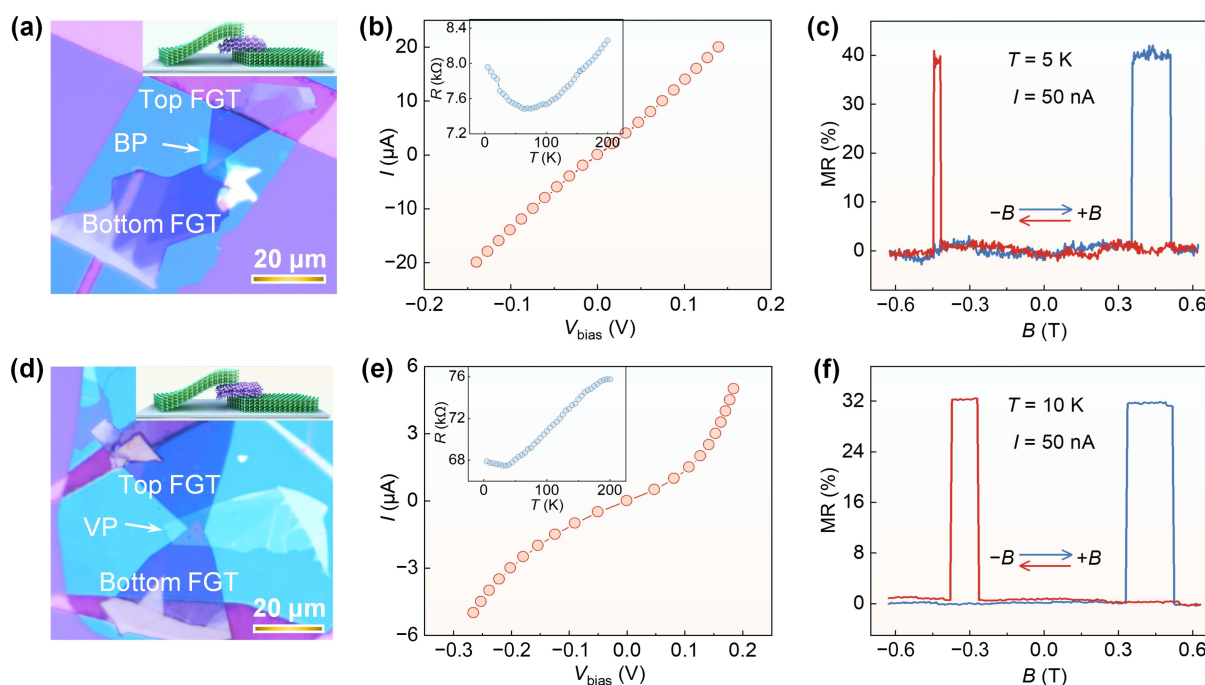


Figure 2 Electrical and magnetotransport properties of the FGT/BP(VP)/FGT heterostructures. (a) Optical image of FGT/BP/FGT heterostructure. Inset: schematic diagrams. (b) I - V curve of FGT/BP/FGT heterostructure measured at 10 K. Inset: temperature dependence of resistance. (c) MR versus OOP magnetic field curve at 5 K (bias current: 50 nA). (d) Optical image of FGT/VP/FGT heterostructure. Inset: schematic diagrams. (e) I - V curve of FGT/VP/FGT heterostructure measured at 10 K. Inset: temperature dependence of resistance. (f) MR versus OOP field curve at 10 K (bias current: 50 nA).

of 50 nA, we observe a maximum MR ratio of 40.4% followed by a decrease trend as the bias current increases to 10 μ A in heterostructure with BP spacer (Fig. 3(c)). Moreover, with increasing bias current, MR ratio exhibits an overall decreasing trend in the heterostructure with VP spacer. In the FGT/BP/FGT and FGT/VP/FGT heterostructures, bias current dependence on MR ratio is primarily attributed to the higher-energy electrons being scattered by localized trap states within the BP (VP) layers or at the interface. To further understand the temperature dependence of MR, we also performed the magnetotransport measurements at the temperature range from 10 to 180 K (Fig. 3(b)). As shown in Fig. 3(d), an almost monotonic decline in MR is observed with the temperature for both FGT/BP/FGT and FGT/VP/FGT heterostructures, originating from the decrease of the spin polarization of FGT with temperature increasing. Based on the Julliere model [7], the spin polarization P of FGT can be calculated by Eq. (2)

$$MR = \frac{P^2}{1 - P^2} \quad (2)$$

The estimated large P are 36.9% (MR = 31.5%, FGT/VP/FGT) and 30.6% (MR = 20.6%, FGT/BP/FGT) at 10 K, which are larger than reported for FGT/MoS₂/FGT (MR = 3.1%, P = 12.3%, 10 K) [28], as shown in Fig. S8 in the ESM. However, compared with other FGT-based heterostructures, such as FGT/InSe/FGT [45] (MR = 41%, P = 41%, 10 K) and FGT/MoSe₂/FGT [46] (MR = 42%, P = 41.6%, 10 K), the maximum P (36.9%) remains lower value. That is to say, choosing the appropriate spacer maybe further improve the spin polarization of FGT-based heterostructures. In addition, using Bloch's law, temperature-dependent spin polarization P can be well fitted by Eq. (3)

$$P(T) = P_0(1 - \alpha T^3) \quad (3)$$

where P_0 and α represent the spin polarization at 0 K and a constant relating to materials, respectively [47]. The α for FGT/VP/FGT and FGT/BP/FGT devices are almost the same value (2.5×10^{-4} and 2.6×10^{-4} K^{-3/2}), which are comparable to values reported in previous studies for FGT-based heterostructures [26, 28, 45].

The resistance-field (R - H) curves for FGT/BP/FGT and FGT/VP/FGT heterostructures were measured at 100 K under 360° field rotation, as shown in Figs. 4(a) and 4(b). When the angle between applied magnetic field and the substrate normal (θ_H) is increased from 0° to 360°, MR shows no angular dependence and remains isotropic for our heterostructures with different spacers (Fig. 4(c)). In contrast, FGT/VP/FGT heterostructure shows higher MR ratio compared to that with BP spacer in the angle range from 0° to 360°, originating from the higher spin polarization in FGT/VP/FGT heterostructure. The H_C for the two FGT layers can reach maximum values in the in-plane (IP) direction ($\theta_H = 90^\circ$ and 270°) (Fig. 4(d) and Fig. S9 in the ESM), while the H_C display very small values in OOP direction ($\theta_H = 0^\circ$ and 180°), indicating strong angular-dependence in the two heterostructures. Additionally, a sharp rise in H_C occurs as the magnetic field approaches IP direction, following a steady increase during its rotation from OOP direction. When the thickness of BP spacer increases to 10 nm, we also observe angular-independent MR and angular-dependent H_C in another fabricated FGT/BP/FGT devices (Fig. S10 in the ESM). In addition, we change the spacer to WSe₂, the similar behaviors (isotropic MR and anisotropic H_C with angle θ_H) are also observed in FGT/WSe₂/FGT heterostructure (Fig. S11 in the ESM). To clarify the origin of the angular-independent MR, we performed angle-dependent AHE measurements on FGT nanoflake at 100 K (Fig. S12(a) in the ESM). As shown in Fig. S12(b) in the ESM, R_{xy} at saturated magnetic field does not change continuously with θ_H , but stays near two stable levels over a broad angular range, with abrupt changes only in narrow angular windows. Based on R_{xy}

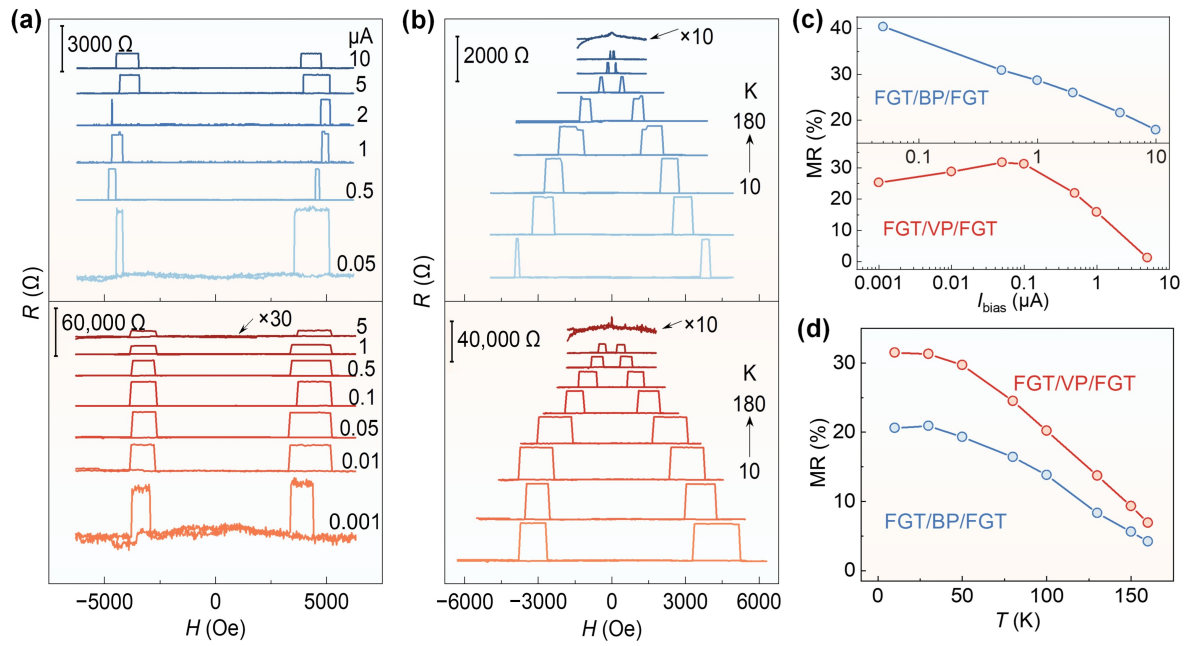


Figure 3 Magneto-transport measurements for FGT/BP(VP)/FGT heterostructures. (a) MR versus OOP magnetic field curves measured with various bias currents for FGT/BP/FGT heterostructure (5 K, top panel) and FGT/VP/FGT heterostructure (10 K, bottom panel). (b) MR versus OOP magnetic field curves measured at the temperature range from 10 to 180 K with a fixed bias current for FGT/BP/FGT heterostructure (5 μ A, top panel) and FGT/VP/FGT heterostructure (50 nA, bottom panel). (c) MR versus bias currents. (d) Temperature dependence of MR. Blue lines and red lines represent BP and VP spacer, respectively.

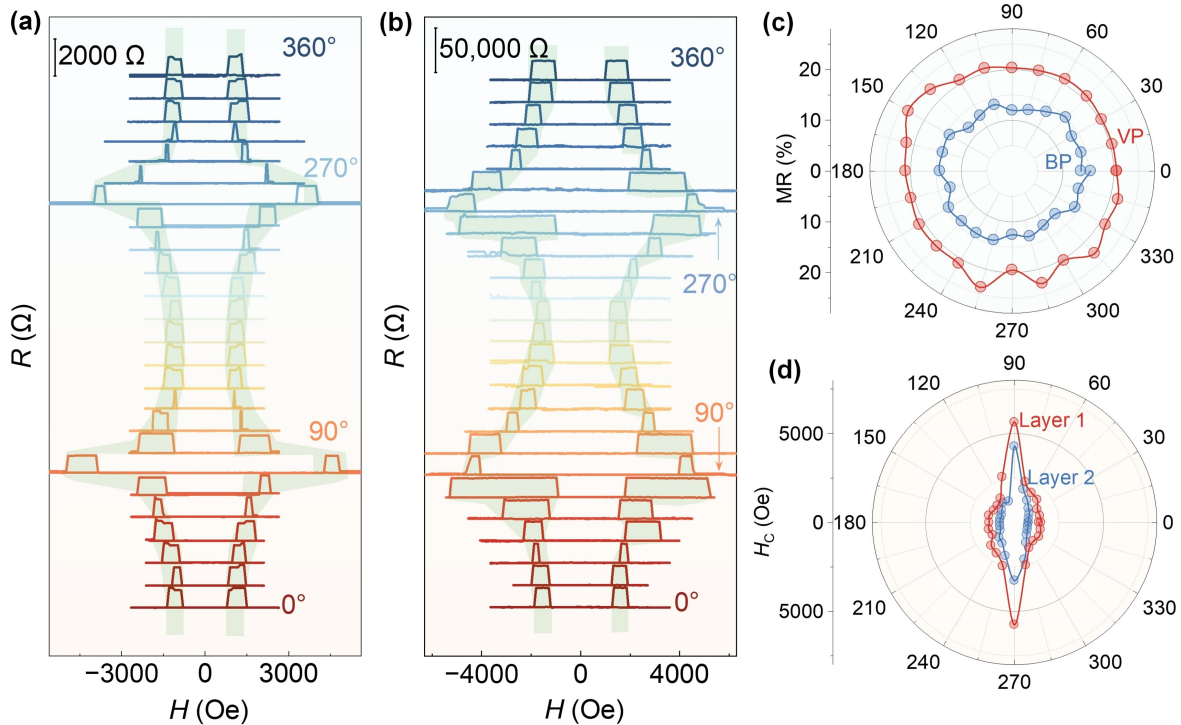


Figure 4 Angular dependence of MR in van der Waals heterostructures at 100 K. (a) and (b) MR as a function of applied magnetic field (R - H) curves for FGT/BP/FGT (a) and FGT/VP/FGT (b), measured at different angles θ_H (θ_H is defined as the angle between applied magnetic field and the substrate normal). (c) Angular dependence of the MR ratio for FGT/BP/FGT (blue line) and FGT/VP/FGT (red line). (d) Polar plot of the coercivities H_c in FGT/BP/FGT.

versus θ_H , magnetization orientation angle (θ_M) can be calculated by Eq. (4) [48]

$$\theta_M = \arccos\left(\frac{R_{xy}(\theta_M)}{R_{xy}(\theta_H = 0)}\right) \quad (4)$$

The extracted $\theta_M - \theta_H$ relation shows that the magnetization

remains close to two out-of-plane orientations and switches abruptly between them, instead of continuously rotating toward the field direction, as shown in Fig. S12(c) in the ESM. This behavior reflects the strong PMA of FGT. The magnetic field mainly drives transitions between bistable perpendicular magnetic configurations, and the magnetization reversal may be dominated by domain wall

motion rather than coherent rotation, which is consistent with previous work on domain wall motion in vdW ferromagnet FGT [44]. Since the MR is governed by the relative magnetization configuration of the two ferromagnetic electrodes, such discrete switching keeps the heterostructures predominantly in parallel or antiparallel states over most field angles. The properties of angular-independent MR and angular-dependent H_C in the FGT-based heterostructure maybe originate from PMA and domain-wall-motion-mediated magnetization reversal of FGT.

In FGT-based vdW spintronic heterostructures, large MR ratios of $\sim 160\%$ and $\sim 120\%$ have been reported at 4.2 K for devices with h -BN [26] and WSe_2 [49] spacers, respectively. At the temperature of 10 K, our FGT/monoelemental phosphorus heterostructures show comparable MR ratios (40.4% for BP and 31.5% for VP) (Fig. 5(a)), compared with other heterostructures with different spacers [28–30, 45, 46]. To further analyze higher MR ratios in our heterostructures, the spin-dependent transport properties were calculated within the framework of density functional theory (DFT) combined with the non-equilibrium Green's function (NEGF) formalism, as implemented in the Nanodcal software package [50]. The atomic structure of FGT/BP/FGT is shown in Fig. 5(b). The spin-summed transmission coefficients for P and AP alignments of the magnetization direction in the top and bottom FGT layers show energy-dependent behaviors (upper panel of Fig. 5(c)). Moreover, compared with the parallel state, lower transmission coefficients of

AP state in the entire energy range indicates a higher-resistance regime, which is consistent with our experimental results. Based on these transmission coefficients, the calculated MR ratio in the heterostructure reaches the maximum of 89,104% at 0.32 eV below the Fermi level, and it is about 502% at the Fermi level (lower panel of Fig. 5(c)), implying the potential application of the heterostructure in the spintronics.

4 Conclusions

In summary, we report a magnetoresistive spin valve effect in vdW heterostructures of FGT/BP/FGT and FGT/VP/FGT. The observed nonlinear I - V curves demonstrate the tunneling barrier behavior of BP/VP in the heterostructures. The FGT/BP/FGT and FGT/VP/FGT heterostructures exhibit MR ratios of 40.4% and 31.5%, respectively. The MR ratio exhibits a strong dependence on bias current and thickness of monoelemental phosphorus tunnel barrier. Interestingly, our FGT-based heterostructures exhibit an isotropic MR regardless of applied magnetic field orientation, originating from PMA and domain-wall-motion-mediated magnetization reversal of FGT with respect to the applied field, which realizes the development of robust memory devices enough to function in arbitrary or uncontrolled magnetic environments. This study paves the way for the development of novel spintronic devices with magnetic field angular-independent MR.

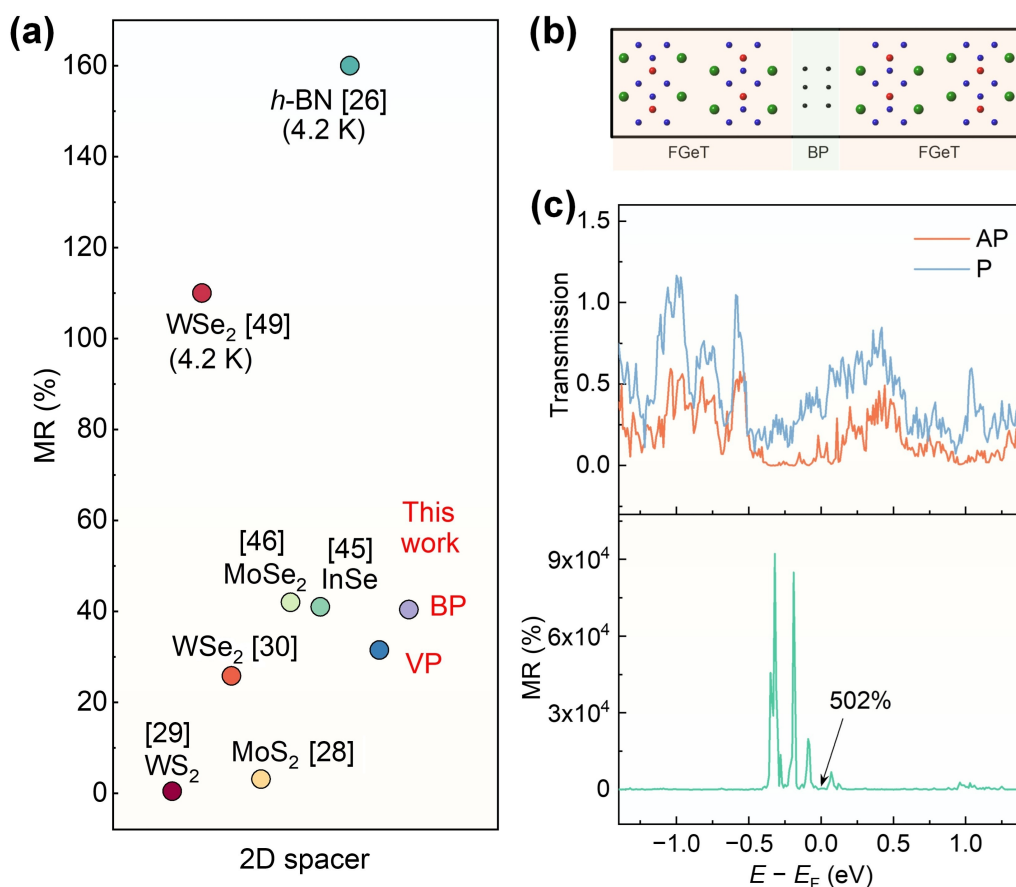


Figure 5 Comparison of MR for all-vdW FGT/spacer/FGT heterostructures from previous reports and first-principles calculation of spin-dependent transport properties in FGT/BP/FGT heterostructure. (a) Benchmark comparison of MR ratios for different FGT/spacer/FGT devices at low temperature (4.2 K for h -BN [26] and WSe_2 [49], 10 K for MoS_2 [28], WS_2 [29], InSe [45], MoSe_2 [46] and WSe_2 [30]). Data from this work is highlighted with brown arrows. (b) Atomic configuration of FGT/BP/FGT heterostructure. (c) Spin-summed transmission coefficients for parallel and antiparallel alignments of the magnetization direction in the top and bottom FGT layers, and the derived MR ratio as a function of electron energy (Fermi level = 0).

Electronic Supplementary Material: Supplementary material (experimental and characterizations details) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94908859>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

Acknowledgements

This work was supported by the National Natural Science Foundation of China (Nos. U24A6002 to Y. J., 52571263 to Z. Y. J., and 52571229 to K. Y.), the National Key Research and Development Program of China (No. 2022YFA1204000 to Y. J.), and the Tianjin Key Research and Development Program (No. 24YFXTZH00150 to Z. Y. J.). Additional support was provided by the Young Top-Notch Talent Program of the Hebei Education Department Science and Technology Project (No. BJK2023082 to Z. Y. J.). Y. J. and Z. Y. J. also acknowledge research support from the Tianjin Municipal Science and Technology Bureau. We would like to thank the Analytical & Testing Center at Tiangong University for AFM work.

Declaration of competing interest

All the contributing author reports no conflict of interests in this work.

Author contribution statement

Z. Y. J., F. Z., and Y. J. conceived and supervised the research. Y. X. T., K. Y., and Y. W. fabricated the devices, analyzed the data, and drafted the manuscript. X. Q. G. and X. Y. G. synthesized FGT single crystals. Y. X. T., Y. H. S., and M. F. Z. performed measurements. Z. Y. J. and F. Z. supervised the manuscript. All authors discussed the results and commented on the manuscript.

Use of AI statement

None.

References

- [1] Iqbal, M. Z.; Iqbal, M. W.; Lee, J. H.; Kim, Y. S.; Chun, S. H.; Eom, J. Spin valve effect of NiFe/graphene/NiFe junctions. *Nano Res.* **2013**, *6*, 373–380.
- [2] Jiang, C. P.; Li, J. H.; Wang, C.; Lu, S. Y.; Yao, X. Y.; Cai, W. L.; Xiong, D. R.; Fan, X. F.; Liu, H. X.; Wang, G. F. et al. Unveiling the endurance limit of SOT-MRAM with CoFeB/MgO/CoFeB junctions: From elemental interdiffusion to array-level reliability. *Nano Res.* **2026**, *19*, 94908447.
- [3] Gong, F.; Deng, W. J.; Wu, Y.; Liu, F. M.; Guo, Y. H.; Che, Z. L.; Li, J. J.; Li, J. Z.; Chai, Y.; Zhang, Y. Z. Reconfigurable logic and in-sensor encryption operations in an asymmetrically tunable van der Waals heterostructure. *Nano Res.* **2024**, *17*, 3113–3119.
- [4] Dieny, B.; Prejbeanu, I. L.; Garello, K.; Gambardella, P.; Freitas, P.; Lehdorff, R.; Raberg, W.; Ebels, U.; Demokritov, S. O.; Akerman, J. Opportunities and challenges for spintronics in the microelectronics industry. *Nat. Electron.* **2020**, *3*, 446–459.
- [5] Dieny, B.; Speriosu, V. S.; Parkin, S. S. P.; Gurney, B. A.; Wilhoit, D. R.; Mauri, D. Giant magnetoresistance in soft ferromagnetic multilayers. *Phys. Rev. B* **1991**, *43*, 1297–1300.
- [6] Anguelouch, A.; Schrag, B. D.; Xiao, G.; Lu, Y.; Trouilloud, P. L.; Wanner, R. A.; Gallagher, W. J.; Parkin, S. S. P. Two-dimensional magnetic switching of micron-size films in magnetic tunnel junctions. *Appl. Phys. Lett.* **2000**, *76*, 622–624.
- [7] Julliere, M. Tunneling between ferromagnetic films. *Phys. Lett. A* **1975**, *54*, 225–226.
- [8] Moodera, J. S.; Kinder, L. R.; Wong, T. M.; Meservey, R. Large magnetoresistance at room temperature in ferromagnetic thin film tunnel junctions. *Phys. Rev. Lett.* **1995**, *74*, 3273–3276.
- [9] Piquemal-Banci, M.; Galceran, R.; Martin, M. B.; Godel, F.; Anane, A.; Petroff, F.; Dlubak, B.; Seneor, P. 2D-MTJs: Introducing 2D materials in magnetic tunnel junctions. *J. Phys. D: Appl. Phys.* **2017**, *50*, 203002.
- [10] Zhang, L. S.; Zhou, J.; Li, H.; Shen, L.; Feng, Y. P. Recent progress and challenges in magnetic tunnel junctions with 2D materials for spintronic applications. *Appl. Phys. Rev.* **2021**, *8*, 021308.
- [11] Sierra, J. F.; Fabian, J.; Kawakami, R. K.; Roche, S.; Valenzuela, S. O. Van der Waals heterostructures for spintronics and opto-spintronics. *Nat. Nanotechnol.* **2021**, *16*, 856–868.
- [12] Coy Diaz, H.; Avila, J.; Chen, C. Y.; Addou, R.; Asensio, M. C.; Batzill, M. Direct observation of interlayer hybridization and Dirac relativistic carriers in graphene/MoS₂ van der Waals heterostructures. *Nano Lett.* **2015**, *15*, 1135–1140.
- [13] Roemer, R.; Lee, D. H. D.; Smit, S.; Zhang, X.; Godin, S.; Hamza, V.; Jian, T.; Larkin, J.; Shin, H.; Liu, C. et al. Unraveling the electronic structure and magnetic transition evolution across monolayer, bilayer, and multilayer ferromagnetic Fe₃GeTe₂. *npj 2D Mater. Appl.* **2024**, *8*, 63.
- [14] Zhang, Y.; Lu, H. Y.; Zhu, X. G.; Tan, S. Y.; Feng, W.; Liu, Q.; Zhang, W.; Chen, Q. Y.; Liu, Y.; Luo, X. B. et al. Emergence of Kondo lattice behavior in a van der Waals itinerant ferromagnet, Fe₃GeTe₂. *Sci. Adv.* **2018**, *4*, eaao6791.
- [15] Zhuang, H. L.; Kent, P. R. C.; Hennig, R. G. Strong anisotropy and magnetostriction in the two-dimensional Stoner ferromagnet Fe₃GeTe₂. *Phys. Rev. B* **2016**, *93*, 134407.
- [16] León-Brito, N.; Bauer, E. D.; Ronning, F.; Thompson, J. D.; Movshovich, R. Magnetic microstructure and magnetic properties of uniaxial itinerant ferromagnet Fe₃GeTe₂. *J. Appl. Phys.* **2016**, *120*, 083903.
- [17] Fei, Z. Y.; Huang, B.; Malinowski, P.; Wang, W. B.; Song, T. C.; Sanchez, J.; Yao, W.; Xiao, D.; Zhu, X. Y.; May, A. F. et al. Two-dimensional itinerant ferromagnetism in atomically thin Fe₃GeTe₂. *Nat. Mater.* **2018**, *17*, 778–782.
- [18] Tan, C.; Lee, J.; Jung, S. G.; Park, T.; Albarakati, S.; Partridge, J.; Field, M. R.; McCulloch, D. G.; Wang, L.; Lee, C. Hard magnetic properties in nanoflake van der Waals Fe₃GeTe₂. *Nat. Commun.* **2018**, *9*, 1554.
- [19] Park, S. Y.; Kim, D. S.; Liu, Y.; Hwang, J.; Kim, Y.; Kim, W.; Kim, J. Y.; Petrovic, C.; Hwang, C.; Mo, S. K. et al. Controlling the magnetic anisotropy of the van der Waals ferromagnet Fe₃GeTe₂ through hole doping. *Nano Lett.* **2020**, *20*, 95–100.
- [20] Deng, Y. J.; Yu, Y. J.; Song, Y. C.; Zhang, J. Z.; Wang, N. Z.; Sun, Z. Y.; Yi, Y. F.; Wu, Y. Z.; Wu, S. W.; Zhu, J. Y. et al. Gate-tunable room-temperature ferromagnetism in two-dimensional Fe₃GeTe₂. *Nature* **2018**, *563*, 94–99.
- [21] Roemer, R.; Liu, C.; Zou, K. Robust ferromagnetism in wafer-scale monolayer and multilayer Fe₃GeTe₂. *npj 2D Mater. Appl.* **2020**, *4*, 33.
- [22] Chen, B.; Yang, J. H.; Wang, H. D.; Imai, M.; Ohta, H.; Michioka, C.; Yoshimura, K.; Fang, M. H. Magnetic properties of layered itinerant electron ferromagnet Fe₃GeTe₂. *J. Phys. Soc. Japan* **2013**, *82*, 124711.
- [23] May, A. F.; Calder, S.; Cantoni, C.; Cao, H. B.; McGuire, M. A. Magnetic structure and phase stability of the van der Waals bonded

- ferromagnet Fe_3GeTe_2 . *Phys. Rev. B* **2016**, 93, 014411.
- [24] Yi, J. Y.; Zhuang, H. L.; Zou, Q.; Wu, Z. M.; Cao, G. X.; Tang, S. W.; Calder, S. A.; Kent, P. R. C.; Mandrus, D.; Gai, Z. Competing antiferromagnetism in a quasi-2D itinerant ferromagnet: Fe_3GeTe_2 . *2D Mater.* **2017**, 4, 011005.
- [25] Liu, B. J.; Zou, Y. M.; Zhou, S. M.; Zhang, L.; Wang, Z.; Li, H. X.; Qu, Z.; Zhang, Y. H. Critical behavior of the van der Waals bonded high T_C ferromagnet Fe_3GeTe_2 . *Sci. Rep.* **2017**, 7, 6184.
- [26] Wang, Z.; Sapkota, D.; Taniguchi, T.; Watanabe, K.; Mandrus, D.; Morpurgo, A. F. Tunneling spin valves based on $\text{Fe}_3\text{GeTe}_2/\text{hBN}/\text{Fe}_3\text{GeTe}_2$ van der Waals heterostructures. *Nano Lett.* **2018**, 18, 4303–4308.
- [27] Ikeda, S.; Miura, K.; Yamamoto, H.; Mizunuma, K.; Gan, H. D.; Endo, M.; Kanai, S.; Hayakawa, J.; Matsukura, F.; Ohno, H. A perpendicular-anisotropy CoFeB/MgO magnetic tunnel junction. *Nat. Mater.* **2010**, 9, 721–724.
- [28] Lin, H. L.; Yan, F. G.; Hu, C.; Lv, Q. S.; Zhu, W. K.; Wang, Z. A.; Wei, Z. M.; Chang, K.; Wang, K. Y. Spin-valve effect in $\text{Fe}_3\text{GeTe}_2/\text{MoS}_2/\text{Fe}_3\text{GeTe}_2$ van der Waals heterostructures. *ACS Appl. Mater. Interfaces* **2020**, 12, 43921–43926.
- [29] Hu, C.; Yan, F. G.; Li, Y. C.; Wang, K. Y. Vertical WS_2 spin valve with Ohmic property based on Fe_3GeTe_2 electrodes. *Chin. Phys. B* **2021**, 30, 097505.
- [30] Zheng, Y. H.; Ma, X. L.; Yan, F. G.; Lin, H. L.; Zhu, W. K.; Ji, Y.; Wang, R. S.; Wang, K. Y. Spin filtering effect in all-van der Waals heterostructures with WSe_2 barriers. *npj 2D Mater. Appl.* **2022**, 6, 62.
- [31] Ba, K.; Jiang, W.; Cheng, J. X.; Bao, J. X.; Xuan, N. N.; Sun, Y. Y.; Liu, B.; Xie, A. Z.; Wu, S. W.; Sun, Z. Z. Chemical and bandgap engineering in monolayer hexagonal boron nitride. *Sci. Rep.* **2017**, 7, 45584.
- [32] Kim, S.; Myeong, G.; Shin, W.; Lim, H.; Kim, B.; Jin, T.; Chang, S.; Watanabe, K.; Taniguchi, T.; Cho, S. Thickness-controlled black phosphorus tunnel field-effect transistor for low-power switches. *Nat. Nanotechnol.* **2020**, 15, 203–206.
- [33] Li, C. X.; Xu, X. G.; Ma, Y. L.; Zhao, B.; Zhang, T. Z.; Wang, L.; Wu, Y.; Meng, K. K.; Chen, J. K.; Jiang, Y. Room-temperature non-local spin transport in few-layer black phosphorus passivated with MgO . *Adv. Elect. Mater.* **2022**, 8, 2101048.
- [34] Xu, L. L.; Feng, J. F.; Zhao, K. K.; Lv, W. M.; Han, X. F.; Liu, Z. Y.; Xu, X. H.; Huang, H.; Zeng, Z. M. Magnetoresistance effect in $\text{NiFe}/\text{BP}/\text{NiFe}$ vertical spin valve devices. *Adv. Condens. Matter Phys.* **2017**, 2017, 9042823.
- [35] Galbiati, M.; Dubois, S. M. M.; Wei, H.; Zlatko, V.; Peiro, J.; Godel, F.; Galceran, R.; Brus, P.; Fossard, F.; Mérot, J. S. et al. Unveiling spin selective mechanism toward very large spin signals in black phosphorus 2D semiconductor spintronics devices. *ACS Appl. Mater. Interfaces* **2025**, 17, 50833–50841.
- [36] Sun, Z. Y.; Cai, Z. H.; Cheng, P.; Chen, L.; Zhao, X. W.; Zhang, J. Y.; Wu, K. H.; Feng, B. J. Bound excitons and bandgap engineering in violet phosphorus. *npj 2D Mater. Appl.* **2023**, 7, 37.
- [37] Li, G. H.; Ma, S. P.; Li, Z.; Zhang, Y. W.; Diao, J. L.; Xia, L.; Zhang, Z. W.; Huang, Y. High-quality ferromagnet Fe_3GeTe_2 for high-efficiency electromagnetic wave absorption and shielding with wideband radar cross section reduction. *ACS Nano* **2022**, 16, 7861–7879.
- [38] Liang, S.; Hasan, M. N.; Seo, J. H. Direct observation of Raman spectra in black phosphorus under uniaxial strain conditions. *Nanomaterials* **2019**, 9, 566.
- [39] Zhang, L. H.; Huang, H. Y.; Lv, Z. X.; Li, L. R.; Gu, M. Y.; Zhao, X. W.; Zhang, B.; Cheng, Y. H.; Zhang, J. Y. Phonon properties of bulk violet phosphorus single crystals: Temperature and pressure evolution. *ACS Appl. Electron. Mater.* **2021**, 3, 1043–1049.
- [40] Dolui, K.; Narayan, A.; Rungger, I.; Sanvito, S. Efficient spin injection and giant magnetoresistance in $\text{Fe}/\text{MoS}_2/\text{Fe}$ junctions. *Phys. Rev. B* **2014**, 90, 041401.
- [41] Liu, L.; Wang, D.; Wang, D. D.; Sun, Y.; Lin, H.; Gong, X. L.; Zhang, Y. F.; Tang, R. F.; Mai, Z. H.; Hou, Z. P. et al. Domain wall magnetic tunnel junction-based artificial synapses and neurons for all-spin neuromorphic hardware. *Nat. Commun.* **2024**, 15, 4534.
- [42] Urdampilleta, M.; Klyatskaya, S.; Cleuziou, J. P.; Ruben, M.; Wernsdorfer, W. Supramolecular spin valves. *Nat. Mater.* **2011**, 10, 502–506.
- [43] Xiong, Z. H.; Wu, D.; Vally Vardeny, Z.; Shi, J. Giant magnetoresistance in organic spin-valves. *Nature* **2004**, 427, 821–824.
- [44] Zhang, W. J.; Ma, T. P.; Hazra, B. K.; Meyerheim, H.; Rigvedi, P.; Yin, Z. H.; Srivastava, A. K.; Wang, Z.; Gu, K.; Zhou, S. M. et al. Current-induced domain wall motion in a van der Waals ferromagnet Fe_3GeTe_2 . *Nat. Commun.* **2024**, 15, 4851.
- [45] Zhu, W. K.; Lin, H. L.; Yan, F. G.; Hu, C.; Wang, Z. A.; Zhao, L. X.; Deng, Y. C.; Kudrynskyi, Z. R.; Zhou, T.; Kovalyuk, Z. D. et al. Large tunneling magnetoresistance in van der Waals ferromagnet/semiconductor heterojunctions. *Adv. Mater.* **2021**, 33, 2104658.
- [46] Zhu, S. G.; Lin, H. L.; Zhu, W. K.; Li, W. H.; Zhang, J.; Wang, K. Y. Voltage tunable sign inversion of magnetoresistance in van der Waals $\text{Fe}_3\text{GeTe}_2/\text{MoSe}_2/\text{Fe}_3\text{GeTe}_2$ tunnel junctions. *Appl. Phys. Lett.* **2024**, 124, 222401.
- [47] Shang, C. H.; Nowak, J.; Jansen, R.; Moodera, J. S. Temperature dependence of magnetoresistance and surface magnetization in ferromagnetic tunnel junctions. *Phys. Rev. B* **1998**, 58, R2917–R2920.
- [48] Tang, M.; Huang, J. W.; Qin, F.; Zhai, K.; Ideue, T.; Li, Z. Y.; Meng, F. H.; Nie, A. M.; Wu, L. L.; Bi, X. Y. et al. Continuous manipulation of magnetic anisotropy in a van der Waals ferromagnet via electrical gating. *Nat. Electron.* **2023**, 6, 28–36.
- [49] Min, K. H.; Lee, D. H.; Choi, S. J.; Lee, I. H.; Seo, J.; Kim, D. W.; Ko, K. T.; Watanabe, K.; Taniguchi, T.; Ha, D. H. et al. Tunable spin injection and detection across a van der Waals interface. *Nat. Mater.* **2022**, 21, 1144–1149.
- [50] Taylor, J.; Guo, H.; Wang, J. *Ab initio* modeling of quantum transport properties of molecular electronic devices. *Phys. Rev. B* **2001**, 63, 245407.



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