

Electronic Supplementary Material

High current limits in chemical vapor deposited graphene spintronic devices

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1. Raman spectroscopy of CVD graphene

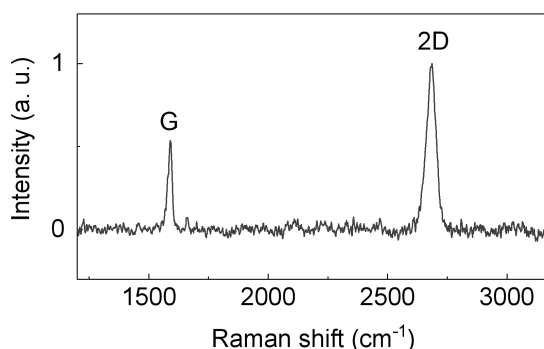


Figure S1 Raman spectra of CVD graphene on Si/SiO₂ substrate used in experiments.

To understand the quality of graphene, we performed Raman spectroscopy of the CVD graphene sample on Si/SiO₂ substrate under 532 nm laser excitation. The 2D to G intensity ratio is ~ 2 which suggests good quality graphene with no defect peaks (Fig. S1).

2. Low current device characterization

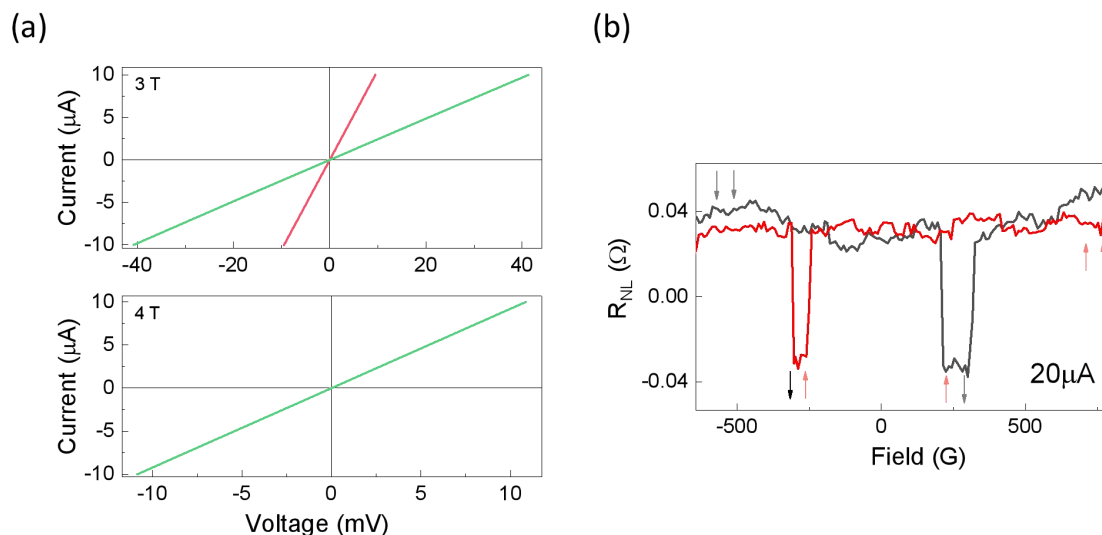


Figure S2 (a) I-V curves measured in 3T configuration to obtain the contact resistance of the injector and detector and 4T I-V curves to calculate the resistivity of the graphene sheet. (b) Spin transport signal measured in the spin-valve geometry showing switching between parallel ($\uparrow\uparrow$ or $\downarrow\downarrow$) to antiparallel ($\downarrow\uparrow$ or $\uparrow\downarrow$) configurations of the injector and detector electrodes at current $I = 20 \mu\text{A}$.

We measured the contact resistance of the ferromagnetic tunnel contacts using 3T configuration I-V characteristics (Fig. S2a). The contact resistance was found to be in the range of 1 - 4 k Ω . To get an efficient spin injection into the graphene the contact resistance should be higher than the graphene sheet resistance. We have also performed four-terminal I-V characteristics to calculate the graphene sheet resistance (Fig. S2a) and observed the sheet resistance of the graphene is 540 Ωsq^{-1} .

We performed nonlocal (NL) measurements to observe spin transport. In the NL scheme, spin currents are electrically injected into the graphene channel through the current circuit (I), and the resulting diffusion spin current at a distance of L is detected by the voltage circuit (V). Such isolation of the current and voltage circuits allows for faithful measurement of pure spin accumulation, eliminating spurious magneto-resistive contributions associated with direct charge currents. By sweeping an in-plane magnetic field ($B_{||}$), the relative magnetic orientations of the injector and detector electrodes are switched from parallel to antiparallel configuration, which results in a spin-valve switching in the measured nonlocal spin voltage ΔV ($\Delta V_{NL} = V_{NL}(\uparrow\uparrow) - V_{NL}(\downarrow\uparrow)$). As depicted in the spin chemical potential profile, this allows for the measurement of the difference in spin chemical potentials for up ($\mu\uparrow$) and down spins ($\mu\downarrow$), also known as the net spin accumulation at a distance L. In our experiments, we perform NL measurements by sweeping the magnetic field forward (-B to +B) and reverse (+B to -B). A clear spin valve signal with spin NL spin-valve resistance $\Delta R_{NL} = \Delta V_{NL}/I \sim 68 \text{ m}\Omega$ was observed at room temperature (shown in Fig. S2b). The obtained values are in nearly the same order as previously reported on Si/SiO₂ devices [1–3].

3. High current T and 4T I-V characteristics

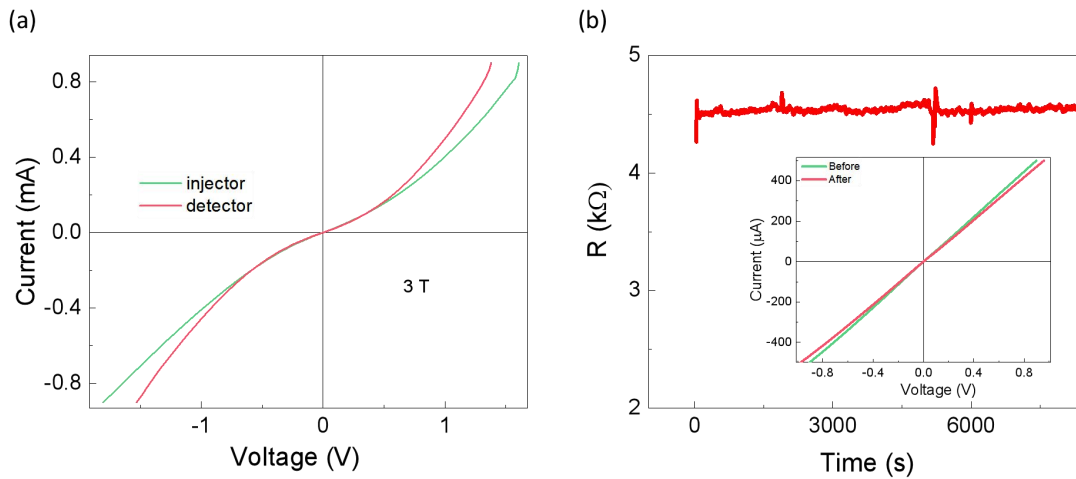


Figure S3 (a) The contact resistance of the injector and detector measured in a 3-terminal (3T) configuration at a higher bias current. (b) The 4T resistance of the CVD graphene as a function of time measured at a higher bias current of 1 mA and the inset shows the four-probe I-V curves before and after endurance measurements.

We have measured the contact resistance of the tunnel contact using the 3T configuration in the device at a higher bias current (Fig. S3a). The 3T I-V clearly shows the nonlinear behavior at a higher bias current also, expected for tunnel contacts used in graphene spin valves. The nonlinearity in I-V curves was observed due to the tunneling behavior across the junction as well as the self-heating at higher bias voltage [4–6] and some variations in resistance can be associated with inhomogeneous graphene/contact interface or nanofilaments formation. For these specific contacts, the contacts show low resistance ($\sim \text{k}\Omega$), and the contact shows non-linear tunneling characteristics for fairly large currents without any degradation. Figure S3b shows the resistance of the graphene as a function of time for applied constant bias current of 1 mA. After a long time (about 9000 s), the graphene still has sustained nearly the same resistance with a higher bias current.

References

- [1] L. Banszerus, M. Schmitz, S. Engels, M. Goldsche, K. Watanabe, T. Taniguchi, B. Beschoten, and C. Stampfer, *Ballistic Transport Exceeding 28 Mm in CVD Grown Graphene*, Nano Lett. **16**, 1387 (2016).
- [2] K. Verguts, Y. Defossez, A. Leonhardt, J. De Messemaeker, K. Schouteden, C. Van Haesendonck, C. Huyghebaert, S. De Gendt, and S. Brems, *Growth of Millimeter-Sized Graphene Single Crystals on Al₂O₃ (0001)/Pt(111) Template Wafers Using Chemical Vapor Deposition*, ECS J. Solid State Sci. Technol. **7**, M195 (2018).

- [3] M. V. Kamalakar, C. Groenveld, A. Dankert, and S. P. Dash, *Long Distance Spin Communication in Chemical Vapour Deposited Graphene*, Nat. Commun. **6**, 6766 (2015).
- [4] T. Yu, E. K. Lee, B. Briggs, B. Nagabhirava, and B. Yu, *Bilayer Graphene System: Current-Induced Reliability Limit*, IEEE Electron Device Lett. **31**, 1155 (2010).
- [5] K. J. Lee, A. P. Chandrakasan, and J. Kong, *Breakdown Current Density of CVD-Grown Multilayer Graphene Interconnects*, IEEE Electron Device Lett. **32**, 557 (2011).
- [6] R. Murali, Y. Yang, K. Brenner, T. Beck, and J. D. Meindl, *Breakdown Current Density of Graphene Nanoribbons*, Appl. Phys. Lett. **94**, 2007 (2009).