We demonstrate electrical spin injection from a ferromagnet to a bilayer graphene (BLG) through a monolayer (ML) of single-crystal hexagonal boron nitride (h-BN). A Ni$_8$Fe$_{19}$/ML h-BN/BLG/h-BN structure is fabricated using a micromechanical cleavage and dry transfer technique. The transport properties across the ML h-BN layer exhibit tunnel barrier characteristics. Spin injection into BLG has been detected through non local magnetoresistance measurements.

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structure, BLG and a ML of h-BN is fabricated using a micromechanical cleavage and dry transfer method.\textsuperscript{50,25} The thicknesses of the graphene and h-BN layers as measured by an atomic force microscopy are 0.70 and 0.34 nm, respectively. These thicknesses correspond to two and one MLs of graphene and h-BN, respectively. Finally, a 30 nm Py layer and a non magnetic 45 nm Au/6 nm Ti electrode are fabricated by standard electron beam (EB) lithography and EB evaporation. The doped Si substrate is used as a back gate for controlling the carrier concentration of the BLG. The width of the BLG is 1 μm and the width of the three ferromagnetic electrodes, Py1, Py2, and Py3, are 270, 580, and 380 nm, respectively. The distance between ferromagnetic electrodes Py2 and Py3 is 600 nm. The mobility of the BLG is determined as 2700 and 2300 cm\(^2\)V\(^{-1}\)s\(^{-1}\) at 30 and 300 K, respectively.

We measured the current–voltage (I–V) characteristics of the NiFe/ML h-BN/BLG junction using three-terminal measurements, i.e., current, is applied from Py2 to Py1 and voltage is measured between Py2 and Au/Ti. The I–V curve at 30 K under various back gate voltages \(V_G\) is shown in Fig. 2(a). A non linear I–V relationship was observed. The shape of the I–V curve weakly depends on \(V_G\) within \(V_G = -50\) to +50 V. After the measurement of several junction resistances, we obtained a resistance area product \(R_A\) of the ML h-BN of 0.8–1.2 kΩμm\(^2\) near the zero bias voltage. The change in differential resistance \((dV/dI)\) with respect to \(I_{DC}\) for ML of the h-BN barrier is shown in Fig. 2(b) and reveals a 50% decrease in resistance at an \(I_{DC}\) of ±50 μA (±30 mV). With increasing temperature, the junction resistance monotonically decreases and the ratio \(R_A\) (1.6 K)/\(R_A\) (300 K) is ~2 for most of the ML h-BN junction. The \(R_A\) seems to scale with the number of h-BN layers, as shown in Fig. 2(c). The \(R_A\) increases about a factor of a hundred from one to two MLs of h-BN. This behavior is quite similar to that reported for a h-BN tunnel barrier with a graphene electrode.\textsuperscript{13} These transport measurements suggest that we successfully fabricated a ML-thick h-BN tunnel barrier between a ferromagnetic layer and BLG.

The non local magnetoresistance (NLMR) was measured to detect spin injection and transport in BLG. The measurement circuit is shown in Fig. 1(a). The in-plane magnetic field dependence of the NLMR measured at 30 and 300 K with \(V_G = +50\) V and \(I_{SD} = +50\) μA is shown in Fig. 3(a). Note that the Dirac point (DP) of the BLG is located at \(V_G = +7\) V. A clear NLMR is observed at both 30 and 300 K. This indicates electrical spin injection from ferromagnet to BLG through a ML-thick h-BN barrier. Since the spin-polarized carriers are injected from both ferromagnetic electrodes Py1 and Py2, the NLMR signal displays multiple resistance steps. The relative alignment between Py1, Py2, and Py3 is indicated by arrows in the figure. We define the amplitude of the NLMR, \(\Delta R_{NL}\), as the difference in resistance between positions A and B in Fig. 2(a). The temperature dependence of \(\Delta R_{NL}\) is shown in Fig. 3(b) measured at three different \(V_G\) values. \(\Delta R_{NL}\) shows a peak at low temperature. A similar feature was observed in previous experiments on SLG and MLG devices, and it is considered to be a consequence of either electron–phonon scattering\textsuperscript{26} or the ferromagnetic contact.\textsuperscript{27} Next, the Hanle effect is measured with magnetic field perpendicular to the sample, and the results are shown in Fig. 3(c). A clear Hanle effect signal is observed, which is additional evidence for electrical...
spin injection into the BLG through the h-BN layer. To eliminate any contribution from the Py1 electrode, we determined the Hanle curve using the following procedure. First, we measured the Hanle effect at the magnetization configuration corresponding to positions A and B in Fig. 2(a). Next, we subtract these two curves and divide the result by 2. The obtained Hanle curve can be analyzed using the following equation:

$$\Delta R_{NL} \propto \frac{1}{\sqrt{4\pi D_s t}} \exp \left( -\frac{L^2}{4D_s t} \right) \times \cos \left( \frac{g \mu_B B t}{h} \right) \exp \left( -\frac{t}{\tau_s} \right) dt,$$

where $D_s$ is the spin diffusion constant, $\tau_s$ is the spin relaxation time, $L$ is the distance between the Py electrodes, $g$ is the electron $g$-factor, and $\mu_B$ is the Bohr magneton.

The spin diffusion length $\lambda_s$ is determined as 1.35 and 1.14 $\mu$m at 30 and 300 K, respectively. Considering these values together with the contact resistance and channel length, our device is within the contact-induced spin relaxation region. In this region, the spin relaxation is dominated by spin absorption at the ferromagnetic electrode and thus makes it difficult for us to perform an accurate evaluation of the spin relaxation in the channel. A more reliable determination of $D_s$ and $\tau_s$ can be accomplished by fabricating devices with a longer channel length. This problem will be addressed in future experiments. In addition, since the resistance of the tunnel barrier scales exponentially with the h-BN thickness, the contact-induced spin relaxation can be eliminated with the current channel length if we use two or three MLs of h-BN as a tunnel barrier.

$\Delta R$ is measured at 30 K under various $V_G$ and $I_{dc}$ values, and the results are shown in Fig. 4(a). For comparison, we plot the $V_G$ dependence of the conductance $\sigma$ in Fig. 4(b). First, $\Delta R$ and $\sigma$ clearly coincide. This phenomenon can be observed when the contact resistance is in the so-called transparent regime. Although the h-BN layer acts as a tunnel barrier for charge, for spins we must compare the junction resistance and the spin resistance of graphene $R_G = \rho_G A_s/W_G$, where $\rho_G$ is the resistance and $W_G$ is the width of BLG. From this expression, the junction resistance is comparable to $R_G$, thus spin absorption effects at the ferromagnetic electrode significantly influence the NLMR. $R_G$ increases toward the DP due to the increase of $\rho_G$. Therefore, there is more spin absorption at the ferromagnetic layer and a lower $\Delta R$.

Next, $I_{dc}$ exhibits a much smaller effect on $\Delta R_{NL}$. This observation also supports the claim that $\Delta R$ is dominated by spin absorption at the ferromagnetic layer, which does not depend on $I_{dc}$. $\Delta R$ increases at positive (negative) $I_{dc}$ at a $V_G$ of $+50$ V ($-50$ V). This increase is due to the effect of carrier drift under the injector ferromagnet. According to this model, carrier drift could suppress the spin absorption in the ferromagnetic electrode. The sign of this effect should be reversed when the carrier type is changed from electron to hole, and this can be seen in Fig. 4(a). We performed measurements from 1.6 to 300 K to change the ratio between the contact resistance and $R_G$. A similar $\Delta R$ vs $V_G$ dependence was observed for all measurement temperatures.

In conclusion, we fabricated a ML crystalline h-BN tunnel barrier using a micromechanical cleavage and dry transfer technique. By this technique, we demonstrated spin injection into BLG through a ML h-BN. The spin injection efficiency is limited by the spin current absorption of the ferromagnet because of the low junction resistance of the ML h-BN barrier. Junction resistance can be increased by using a thicker h-BN layer as a tunnel barrier. Nevertheless, our study revealed that a one atom thick crystalline tunnel barrier could be used for electrical spin injection into graphene. The fabrication method presented here is unique compared to methods found in previous studies on lateral spin valve and MTJ devices. This new method should open up new possibilities for utilizing 2D atomic crystal barriers for spintronics.

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1. K. S. Novoselov, D. Jiang, F. Schedin, T. J. Booth, V. V. Khotkevich, S. V. Morozov, and A. K. Geim: Proc. Natl. Acad. Sci. U.S.A. 102 (2005) 10451.
2. K. S. Novoselov, V. I. Fal’ko, L. Colombo, P. R. Gellert, M. G. Schwab, and K. Kim: Nature 490 (2012) 192.
3. Q. H. Wang, K. Kalantar-Zadeh, A. Kis, J. N. Coleman, and M. S. Strano: Nat. Nanotechnol. 7 (2012) 699.
4. W. Han, K. M. McCready, K. Pi, H. W. Wang, Y. Li, H. Wen, J. R. Chen, and R. K. Kawakami: J. Magn. Magn. Mater. 324 (2012) 369.
5. W. Han and R. K. Kawakami: Phys. Rev. Lett. 107 (2011) 047207.
6. T. Y. Yang, J. Balakrishnan, F. Volmer, A. Avsar, M. Jaiswal, J. Samm, S. R. Ali, A. Pachoud, M. Zeng, M. Popinciuc, G. Güntherodt, B. Beschoten, and B. Özyilmaz: Phys. Rev. Lett. 107 (2011) 047206.
7. N. Tombros, C. Jozza, M. Popinciuc, H. T. Jonkman, and B. J. van Wees: Appl. Phys. Express 6 (2013) 073001.
Nature 448 (2007) 571.
8) W. Han, K. Pi, K. M. McCreary, Y. Li, J. J. I. Wong, A. G. Swartz, and R. K. Kawakami: Phys. Rev. Lett. 105 (2010) 167202.
9) A. Avsar, T.-Y. Yang, S. Bae, J. Balakrishnan, F. Volmer, M. Jaiswal, Z. Yi, S. R. Ali, G. Güntherodt, B. H. Hong, B. Beschoten, and B. Özyilmaz: Nano Lett. 11 (2011) 2363.
10) T. Yamaguchi, S. Masubuchi, K. Iguchi, R. Moriya, and T. Machida: J. Magn. Magn. Mater. 324 (2012) 849.
11) S. S. P. Parkin, C. Kaiser, A. Panchula, P. M. Rice, B. Hughes, M. Samant, and S.-H. Yang: Nat. Mater. 3 (2004) 862.
12) S. Yuasa, T. Nagahama, A. Fukushima, Y. Suzuki, and K. Ando: Nat. Mater. 3 (2004) 868.
13) L. Britnell, R. V. Gorbachev, R. Jalil, B. D. Belle, F. Schedin, M. I. Katsnelson, L. Eaves, S. V. Morozov, A. S. Mayorov, N. M. R. Peres, A. H. Castro Neto, J. Leist, A. K. Geim, L. A. Ponomarenko, and K. S. Novoselov: Nano Lett. 12 (2012) 1707.
14) L. Britnell, R. V. Gorbachev, R. Jalil, B. D. Belle, F. Schedin, A. Mishchenko, T. Georgiou, M. I. Katsnelson, L. Eaves, S. V. Morozov, N. M. R. Peres, J. Leist, A. K. Geim, K. S. Novoselov, and L. A. Ponomarenko: Science 335 (2012) 947.
15) G.-H. Lee, Y.-J. Yu, C. Lee, C. Dean, K. L. Shepard, P. Kim, and J. Hone: Appl. Phys. Lett. 99 (2011) 243114.
16) F. Amet, J. R. Williams, A. G. F. Garcia, M. Yankowitz, K. Watanabe, T. Taniguchi, and D. Goldhaber-Gordon: Phys. Rev. B 85 (2012) 073405.
17) T. Georgiou, R. Jalil, B. D. Belle, L. Britnell, R. V. Gorbachev, S. V. Morozov, Y.-J. Kim, A. Gholinia, S. J. Haigh, O. Makarovsky, L. Eaves, L. A. Ponomarenko, A. K. Geim, K. S. Novoselov, and A. Mishchenko: Nat. Nanotechnol. 8 (2013) 100.
18) M. S. Choi, G.-H. Lee, Y.-J. Yu, D.-Y. Lee, S. H. Lee, P. Kim, J. Hone, and W. J. Yoo: Nat. Commun. 4 (2013) 1624.
19) W. J. Yu, Z. Li, H. Zhou, Y. Chen, Y. Wang, Y. Huang, and X. Duan: Nat. Mater. 12 (2013) 246.
20) C. R. Dean, A. F. Young, I. Meric, C. Lee, L. Wang, S. Sorgenfrei, K. Watanabe, T. Taniguchi, P. Kim, K. L. Shepard, and J. Hone: Nat. Nanotechnol. 5 (2010) 722.
21) A. S. Mayorov, R. V. Gorbachev, S. V. Morozov, L. Britnell, R. Jalil, L. A. Ponomarenko, P. Blake, K. S. Novoselov, K. Watanabe, T. Taniguchi, and A. K. Geim: Nano Lett. 11 (2011) 2359.
22) K. Watanabe, T. Taniguchi, and H. Kanda: Nat. Mater. 3 (2004) 404.
23) O. V. Yazyev and A. Pasquarello: Phys. Rev. B 80 (2009) 035408.
24) V. M. Karpan, P. A. Khomyakov, G. Giovannetti, A. A. Starikov, and P. J. Kelly: Phys. Rev. B 84 (2011) 153406.
25) T. Taychatanapat, K. Watanabe, T. Taniguchi, and P. Jarillo-Herrero: Nat. Phys. 7 (2011) 621.
26) T. Maassen, J. J. van den Berg, N. Ibema, F. Fromm, T. Seyller, R. Yakimova, and B. J. van Wees: Nano Lett. 12 (2012) 1498.
27) T. Yamaguchi, R. Moriya, S. Masubuchi, K. Iguchi, and T. Machida: Jpn. J. Appl. Phys. 52 (2013) 040205.
28) K. Pi, W. Han, K. M. McCreary, A. G. Swartz, Y. Li, and R. K. Kawakami: Phys. Rev. Lett. 104 (2010) 187201.
29) T. Maassen, I. J. Vera-Marun, M. H. D. Guimarães, and B. J. van Wees: Phys. Rev. B 86 (2012) 235408.
30) C. Józsa, M. Popinciuc, N. Tombros, H. T. Jonkman, and B. J. van Wees: Phys. Rev. B 79 (2009) 081402.
31) M. Kameno, Y. Ando, E. Shikoh, T. Shinjo, T. Sasaki, T. Oikawa, Y. Suzuki, T. Suzuki, and M. Shiraishi: Appl. Phys. Lett. 101 (2012) 122413.