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ABSTRACT
The nonlocal spin valve configuration consists of two ferromagnetic and nonmagnetic channels, which is an effective configuration for determining spin injection and accumulation. Here, we report that a reversed nonlocal spin signal was detected by changing the voltage probe configurations in graphene (Py/MgO/graphene/MgO/Py) lateral spin valves. The abnormal reversed spin-dependent nonlocal voltage is attributed to the nonuniform pinhole at the interface of the low-resistance tunnel barrier, which makes the charge current flow through the detection electrode and return to the graphene channel. We demonstrate that the channel-width induced spin-polarized current inhomogeneity significantly contributes to nonlocal resistance. A detailed description and simulated results of the tunnel junctions provide evidence for the reversal of the nonlocal voltage sign induced by the low-resistance tunnel barriers. Our work sheds light on the understanding of the spatial distribution of the spin current and the effect of the tunnel barrier, which are essential for the development of spintronic devices.

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INTRODUCTION
Two-dimensional (2D) graphene with a single atomic layer has recently attracted much attention in spintronics because of its unique electronic transport properties.1–3 Graphene spintronics is of interest due to its high mobility and small spin-orbit interaction attributable to the carbon atoms with low atomic mass, whereby resulting in a long spin relaxation time4,5 and excellent spin transport properties. Recently, due to the extraordinary resilience of graphene, flexible graphene spin valves have also been demonstrated, showing excellent spin diffusion.6 Interestingly, few-layer graphene has a complex electronic structure,7–9 with
energy-momentum relationship comprising both linear and parabolic parts, providing a good platform for investigating the spin relaxation further.\textsuperscript{9,29} Additionally, atomically thin oxide barrier MgO is a useful spin tunnel barrier material for spin injection from a ferromagnet (FM) into graphene.\textsuperscript{29-32} Such tunnel barriers are crucial for efficient spin injection due to solving the conductivity mismatch\textsuperscript{32-34} between the metallic ferromagnet (FM) and graphene. To date, a comprehensive understanding of the spin dephasing during spin injection and the background signal induced by the spatial thickness distribution of oxide barriers remains missing. Similarly, the inhomogeneous spin current distribution in graphene nonlocal spin valve (NLSV) induced by width makes an indispensable contribution to the spin signal.\textsuperscript{1,28} To clarify this phenomenon, in this work, we present a systematic investigation on the injection and detection of the spin current in a lateral ferromagnetic-MgO-graphene-MgO-ferromagnetic spin-valve device with a low resistance tunneling barrier interface. The negative spin signal $\Delta R_{NL}$ in five-layer graphene with the crossed configuration was observed when the graphene width was larger than the spacing (L or D) between the injector and detector. Our experimental and simulated results reveal that the inhomogeneous spin current distributions induced by the low-resistance tunnel barriers are responsible for the inverted spin signal behavior. The observed results open avenues for future investigations of spin-polarization distribution in the graphene layer. Moreover, also, providing a deep understanding of the fundamental physics for spin accumulation in graphene is indispensable for the development of spintronic devices.

**EXPERIMENTAL RESULTS AND DISCUSSIONS**

An NLSV configuration consisting of two FM electrodes with graphene bridged by a gold electrode was designed, and electron beam lithography and lift-off technique were used in the device fabrication process, as shown in Fig. 1(a). A scanning electron microscope (SEM) image of the NLSV configuration is shown in Fig. 1(b). The ferromagnetic (FM) electrodes were prepared for Py (NiFe alloy) via sputtering deposition. The oxygen plasma etching technique controlled the width of the graphene [see Fig. 1(d)]. NiFe/MgO tunnel electrodes on graphene were employed to enhance the efficiency of the spin injection. The spins were electrically injected between electrodes Au1 and FM1 [see Fig. 1(d)], and a nonlocal voltage would be detected between electrodes Au2 and FM2 because of the spin accumulation and diffusion. The NLSV has the advantage of isolating current and voltage routes, which enables the detection of pure spin signals under the without spurious magnetoresistive (MR) contributions.\textsuperscript{24,25,28-30} Also, to control the magnetization of the FM electrodes, we swept an in-plane magnetic field. Thus, we can measure the MR with parallel and antiparallel configurations of FM2 and FM1, resulting in a measurement of the spin splitting ($\Delta\mu$) induced in the graphene channel and the spin polarization of the NiFe/MgO electrodes.\textsuperscript{28-31} Various widths of the FM electrodes ensure distinct coercive force for them.

The nonlocal spin transport measurements were carried out by applying a magnetic field parallel to the FM electrodes. First, the spin-polarized electrons were injected into the graphene channel from the FM1 injector through the MgO interface layer and then accumulated and diffused in the graphene channel. Spin diffusion is usually calculated by the chemical potential ($\mu_1$ and $\mu_\parallel$) of spin. Also, the spin density in graphene increases with a split spin of the chemical potential and decreases exponentially with the spin diffusion length.\textsuperscript{24,25,28-30} We measured the graphene NLSV spin signal as a function of the external magnetic field via two different probe configurations, which are named “half” and “cross,” as illustrated in Fig. 2. The major distinction between both configurations is whether the voltage and current probes are measured on the same side or not, as shown in Figs. 2(a) and 2(b), respectively. Figure 2(c) reveals the NLSV spin signal measured in the “half” configuration with a fixed center-to-center channel length of 3 μm and a graphene width of 10 μm. Two rectangular shaped resistance dips representing the antiparallel magnetic configurations of two FMs were observed when the magnetic field swept from ±400 Oe to ±400 Oe. As shown in Fig. 2(c), the high value of the resistance relates to the parallel magnetization of FM1 and FM2, and the low value of the resistance corresponds to the antiparallel state. The nonlocal spin signal $\Delta R_{NL}$ of 30 mΩ is indicated by the arrow. Interestingly, as depicted in Fig. 2(d), a striking feature that the spin signal is inverted is demonstrated: the resistance is high for the antiparallel state and low for the parallel state of FM1 and FM2. The nonlocal spin signal indicates a negative value $\Delta R_{NL} = -28$ mΩ indicated by the arrow and is a little smaller than the signal in the “half” configuration.

In particular, the standard spin signal indicates that the spin polarization of the FM1 injector and FM2 detector have the same signs. Hence, the observed NLSV signal should be the same based on the one-dimensional (1D) drift-diffusion model.\textsuperscript{36,41} However, all the NLSV devices consist of at least 2D configurations. It is essential to consider the inhomogeneous injection at the interface as well as the spatial distribution of the spin current in a graphene channel. It should be mentioned that some switching field deviations happen between the half and cross configurations. The reason is that the switching field has contributed to the dynamic evolution of the
magnetic domain in contact with the graphene channel in the FM electrodes with a multidomain structure. Moreover, we further measure the graphene NLSV spin signal as a function of the external magnetic field by using four different FM electrodes probe in “half” and “cross” configurations with a fixed center-to-center channel length of 3 μm and a graphene width of 10 μm at 10 K. As shown in Fig. 3, the obtained spin-valve signal is 200 mΩ and −120 mΩ, respectively, for “half” and “cross” configurations. Similarly, the

FIG. 2. Schematic illustration of nonlocal graphene spin valve with the half (a) and cross (b) configurations consisting of two FM electrodes. (c) Nonlocal spin signal \( \Delta R_{NL} \) scans of few-layer graphene spin valves measured at room temperature with the half configuration. The black (red) curve exhibits the nonlocal resistance as the magnetic field is swept from up (down). The nonlocal spin signal \( \Delta R_{NL} \) of 30 mΩ is indicated by the arrow. Inset: The nonlocal spin transport measurement of this device with a spacing of \( L = 3 \mu m \) and graphene width of \( W = 5 \mu m \). (d) Nonlocal MR (\( \Delta R_{MR} \)) scans of few-layer graphene spin valves measured at room temperature with the cross configuration. The nonlocal spin signal indicates a negative value \( \Delta R_{NL} = -28 \) mΩ indicated by the arrow.

FIG. 3. Schematic illustration of nonlocal graphene spin valve with the half (a) and cross (b) configurations consisting of four FM electrodes. (c) Nonlocal spin signal \( \Delta R_{NL} \) scans of few-layer graphene spin valves measured at \( T = 10 K \) with the half configuration. The black (red) curve exhibits the nonlocal resistance as the magnetic field is swept from up (down). The nonlocal spin signal \( \Delta R_{NL} \) of 200 mΩ is indicated by the arrow. Inset: The spin transport measurement of this device with a spacing of \( L = 3 \mu m \) and graphene width of \( W = 10 \mu m \). (d) Nonlocal MR (\( \Delta R_{MR} \)) scans of few-layer graphene spin valves measured at \( T = 10 K \) with the cross configuration. The nonlocal spin signal indicates a negative value \( \Delta R_{NL} = -120 \) mΩ indicated by the arrow.
observed spin signal via different FM electrodes indicates that the spin signal is independent of the variability of the FM electrodes.

To better understand the inversion of the spin-dependent nonlocal voltage, the current-voltage ($I-V$) characteristic of the graphene/MgO/Py junctions was measured at room temperature. Figures 4(a) and 4(b) show the measured current-voltage ($I-V$) characteristic with the different thickness of the MgO layers. We found that the graphene/MgO/Py junction indicates a lower interface resistance (∼366 Ω) than that of a typical tunnel junction which is displayed in Fig. 4(b). In our previous studies for graphene LSVs with Py/MgO/Ag junctions, the amount of oxygen was observed to be decreased in the MgO layer. Such an oxygen vacancy acts as pinhole, decreasing the tunneling barrier height and the resistance at the interface. The characterization of the low-resistance tunnel barriers with an MgO thickness of 1.0 nm in our device indicates that the degree of inhomogeneity results in an inversion of the sign of the spin-dependent nonlocal voltage. For example, it is noteworthy that spin injection does not take place over the entire interface but at the specific contact where the interface resistance is comparatively low. The magnitude of $\Delta R_{NL}$ obtained from NLSV measurement is dependent on whether the locations of the current and voltage probes are on the same side of the graphene channel or not.

One should note that the background nonlocal voltage is of the same order (or smaller than) of the spin-dependent measurement signal and changes by a factor of two between the two configurations. This potentially originates from that the MgO tunnel barrier does not grow perfectly layer-by-layer, and the difference in its thickness may cause some structural defects such as pinholes. In this condition, the spin injection across the Py/MgO/graphene contact is nonuniform, and the local current flow holds a broken symmetry then. The contact resistance at certain points is low enough so that there is a charge current flowing through the detection electrode and back into the channel. This results in the background voltage determined by the positions of the voltage injection electrodes. In order to improve the tunneling barrier, we tried to increase the thickness of the MgO layer to a value of 1.4 nm. Therefore, the evidence of tunnel barrier behavior was observed, as demonstrated in Fig. 4(b). Interestingly, we found that the inversion of the spin-dependent part of the nonlocal signal vanished in such tunnel barrier junction devices.

To further understand the graphene channel width effects on the spin accumulation signal, we symmetrically investigate the graphene length and width depending on the spin signal. Figure 4(c) shows the nonlocal graphene spin signal ($\Delta R_{NL}$) as a function of the spacing between the injector and detector in the “cross” configuration. The width of graphene was fixed at 3 μm, and the magnitude of $\Delta R_{NL}$ for both configurations decreases with channel lengths, which indicates the exponential decay of spin relaxation with increasing

![FIG. 4. The four-probe I-V curve between Py/MgO/Gr interfaces measured with MgO thicknesses of (a) 1.0 nm and (b) 1.4 nm. The measured interface resistance is around 366 Ω and 8.9 × 10^4 Ω, respectively. (c) Nonlocal graphene spin signal $\Delta R_{NL}$ as a function of the spacing between the injector and detector in the “cross” configuration. (d) Nonlocal spin signal $\Delta R_{NL}$ scans of graphene spin valves measured by half and cross configurations when the width of graphene reduces to 1 μm at low-temperature T = 10 K. The disappearance of the negative spin signal indicates that the spatial distribution of spin accumulation was diminished with decreasing graphene width.](image-url)
channel length. On the other hand, it is interesting that the difference in the spin signal between both configurations also decreases with the channel length, which indicates that the inhomogeneous spin accumulation becomes weaker.\textsuperscript{38,39} In Fig. 4(d), as the width of graphene reduces to 1 \( \mu \text{m} \), we found that the negative spin signal completely disappears at the graphene channel, indicating that the space inhomogeneous distribution of the spin accumulation was reduced with small graphene width. We observed that the detection of the voltage probe in the NLSV affects the space inhomogeneous distribution of the spin accumulation was reduced with small graphene width. It was experimentally demonstrated that the location of the voltage probe in the NLSV affects the detection of spin accumulation in a graphene channel with a different graphene width even for a fixed channel length.

Thus, to investigate the deviation, the spatial distribution of the spin current was calculated by a 3D network of the spin-polarization-dependent elements. Calculated spin accumulation with the widths between 1 \( \mu \text{m} \) and 10 \( \mu \text{m} \) was demonstrated in Fig. 5. For the dependencies of the spin signal \( \Delta R_{\text{NL}} \) in the transverse direction, one is the spatial distribution of spin accumulation induced by inhomogeneous spin injection at the point interface,\textsuperscript{28–30,34–36} and the other is the junction size.\textsuperscript{37,38} The effect of the spin resistances of the FM electrode on the spin signal can be justified by assuming that the spin relaxation length of the graphene channel is much greater than the junction size or cross section of the channel. Attributed to small spin-orbit interaction, the spin relaxation length of graphene (>1 \( \mu \text{m} \)) is relatively larger than Au (\( \sim 0.17 \mu \text{m} \)) so that the junction size of the device is smaller than the spin relaxation length of graphene.\textsuperscript{24,25,28–30} Consequently, it is believed that the junction size effect plays an essential role in the channel width effect on \( \Delta R_{\text{NL}} \).

As shown in Fig. 5, the spin current distribution in the vicinity of injector NiFe/MgO/graphene and detector NiFe/MgO/graphene interfaces is very inhomogeneous as the width of graphene increases from 1 \( \mu \text{m} \) to 10 \( \mu \text{m} \). In the neighborhood of the injector, the spin current diffusivity density is uniform. However, in the region far from the injector, the current drifts radically and the spin current, which will be measured at the bottom, flows upstream along the top edge of the graphene channel, and then, these local asymmetry spin currents’ spreading will give a distinct voltage at the FM detector. Such an asymmetry creates a different spin polarization and spin accumulation. It could be caused by the large difference variation in the barrier thickness and the conductivity between graphene and FM electrodes,\textsuperscript{38–41} and the tiny spin diffusion length of NiFe.\textsuperscript{42,43} It is notable that the spin accumulation as calculated is certainly inhomogeneous across the width of the channel, but it does not change in sign. We suggest that a charge current flowing through the detector contact with low contact resistance leads to the sign reversal. A more exotic explanation would be related to some inhomogeneous pinning of the Fermi level at the Py/MgO interface, resulting in a local change of sign in the detection efficiency. In particular, most of the spin current is injected near the base edge of the injector NiFe/MgO/graphene interface. Owing to the asymmetry in spin injection and the asymmetrical placement at the detector FM2, the effective length between the injector and detector of the half configuration is shorter than that of the cross configuration. Hence, there is a smaller spin-valve signal \( \Delta R_{\text{NLS}} \) for the cross configuration. Alternatively, the inverted signal indicates an opposite sign of the injector polarization and detector polarization. A probable negative spin accumulation in one of the NiFe/MgO/graphene junctions induced by the low-resistance tunneling barrier can generate an inverted spin signal. Besides, an inverted spin signal in a graphene device, where the graphene is used as a tunneling layer, has been reported previously, and the reversal was caused by the three conduction channels associated with the gate voltage in graphene. However, it is different from our case.\textsuperscript{44} We acknowledge that the reverse spin signal on the interface needs quantitative and detailed understanding through more experimental and theoretical studies. One note that the detailed mechanism for the inverted spin signal is also subject to further investigation.

**CONCLUSIONS**

In conclusion, we investigated the spatial distribution of spin current in (Py/MgO/graphene/MgO/Py) lateral spin valves with various graphene widths and found that the spin signal strongly depends on the location of the voltage probe and the graphene width effects. The inhomogeneous spin injection at the interface induced by the low-resistance tunnel barriers and the spin accumulation of the spatial distribution in the graphene were taken into account in analyzing the NLSV spin signal. Our experimental and simulated results demonstrate that the current inhomogeneous distributions induced by the low-resistance tunnel barriers are responsible for the inverted spin signal behavior. The observed result shows that the inhomogeneous distribution of the spin current in graphene and
the effect of the tunneling barrier are crucial for the development of spintronic devices.

EXPERIMENTAL METHODS

The graphene flakes with the number of layers (3–5 layer) in this study were prepared by mechanical exfoliation techniques. We fabricated the NSLV graphene devices via micro- and nanofabrication techniques. The electrodes which consist of the bilayer Cr (5 nm)/Au (30 nm) structure were deposited by electron beam evaporation equipment at a base pressure of 1 × 10−8 Torr. The widths of the fabricated Py contacts were varied from 70 nm to 300 nm so that a variation of coercivities can be obtained. Interface cleaning was carried out through an ultraviolet radiation technique before the sample was loaded into the deposition chamber. A 1-nm-thick MgO layer was first grown onto the resist-patterned graphene sample using electron beam evaporation techniques followed by a 40-nm-thick Py layer. Finally, the structure is capped with a 5-nm-thick Al2O3 layer for protection. O3 plasma etching techniques were used to control the sample width. Nonlocal spin valve (NSLV) measurements were performed at room temperature by a standard current-bias lock-in technique with a.c. and a current amplitude of 50 μA and a frequency of 79 Hz.

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The authors declare that they have no competing interests.

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