Inverse Spin Valve Effect in Multilayer Graphene Device

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Abstract. We report the gate-voltage dependence of the spin transport in multilayer graphene (MLG) studied experimentally by the local measurement. The sample consists of a Ni/MLG/Ni junction, where the thickness of the MLG is 9 nm and the spacing of two Ni electrodes is 300 nm. At zero gate voltage, we observed the normal spin valve effect, in which the resistance for the antiparallel alignment of magnetization in ferromagnetic electrodes is larger than that for the parallel alignment. By applying a large gate voltage, on the other hand, the spin valve effect is reversed: the resistance for the antiparallel alignment becomes smaller than that for the parallel alignment. The result is qualitatively interpreted as a quantum interference effect, indicating that the mean free path and the spin relaxation length of the MLG are longer than the electrode spacing (300 nm).

1. Introduction
Spin transport in graphite-based materials such as carbon nanotubes [1–4], single-layer graphene (SLG) [5–7], and multilayer graphene (MLG) [8–11] has attracted considerable attention in recent years because coherent spin transport is expected over a long distance due to the weak spin-orbit and hyperfine interactions in these materials. Gate control of spin conduction in such materials is of high interest from the viewpoint of realizing multi-functional spintronic devices and clarifying the underlying physics. In particular, in view of practical applications, MLG should be superior to SLG because the influence of sample imperfections such as ripples on the spin and charge transports is expected to be smaller in MLG due to its stiffness [12], leading to longer mean free path $l_e$ and spin relaxation length $\lambda_N$, although one needs to take into account the finite screening length of the gate electric field in MLG [13]. In this paper, we explore the gate controllability of the spin transport in MLG. We report the gate voltage dependent spin transport, indicating that the mean free path and the spin relaxation length are longer than the electrode spacing of 300 nm.
Figure 1. (a) Atomic force micrograph of the sample for the local measurement. Configuration and materials of electrodes are indicated. (b) Schematic side view of the sample.

2. Experimental

The samples were fabricated by the standard micromechanical cleavage technique of bulk graphite [14], followed by the electron beam lithography and metal deposition: a layer of MLG about 10 nm thick was exfoliated from a grain of kish graphite and placed on a SiO$_2$/highly doped Si substrate. Two ferromagnetic rectangles (Ni, 40 nm thick) with different aspect ratios were connected to the MLG. Then two normal-metal wires (Cr/Au, 5/100 nm thick) were attached to the Ni rectangles as the measurement leads. Figure 1 shows the atomic force micrograph and a schematic side view of the sample.

We examined more than ten samples with different MLG thicknesses and junction resistances. Here we report the results of a sample with 9 nm thick MLG and a resistance of around 40 Ω (depending on the gate voltage), which is the smallest among all samples we have examined so far. In measurement, the output voltage $V$ was measured using the four-terminal lock-in technique with an excitation current $I$ of 10 µA and a frequency of 119 Hz. The in-plane magnetic field $H$ was applied parallel to the long side of the ferromagnetic electrodes. Different aspect ratios of two Ni rectangles lead to different coercive forces, realizing the parallel and anti-parallel alignment of the magnetization depending on the magnetic field. The back gate voltage was applied through the 300 nm thick SiO$_2$ layer. The sample was immersed in liquid $^4$He and kept at 4.2 K. Thickness and sample geometry were determined with AFM and SEM after transport measurement.

3. Results

Figure 2 shows magnetoresistance (MR) curves measured at different gate voltages, $V_g = -80$, 0, and +80 V. The MR exhibits hysteresis corresponding to the parallel and anti-parallel alignment of the magnetization in ferromagnetic electrodes, as indicated in Fig. 2. Here, the variation of the MR is small and obscure in comparison with the conventional MR curves [6] presumably due to the resistance mismatch [15] at the transparent interface between Ni and MLG and due to the domain structures [16] in the wide Ni electrodes. Notice that at $V_g = 0$ the MR for antiparallel configuration of magnetization is larger than that for parallel configuration, corresponding to the normal spin valve effect commonly observed in ferromagnetic metal (FM)/normal metal/FM systems, while at $V_g = -80$ and 80 V the MR for antiparallel configuration is smaller than that for parallel configuration, i.e., the spin valve effect is reversed.

In order to analyze the effect quantitatively, we define spin induced magnetoresistance (SIMR)
Figure 2. Magnetoresistance measured at $V_g = -80, 0, 80$ V from top to bottom. Solid (dashed) lines are MR in increasing (decreasing) magnetic field. Arrows indicate the magnetization configurations of ferromagnetic electrodes.

Figure 3. (a) Spin induced magnetoresistance (SIMR) and (b) resistance at zero field as a function of gate voltage. The SIMR is normalized so that the maximal value is equal to 1.0.

as

$$SIMR = \int_{-H_c}^{0} [R_{\text{down}}(H) - R_{\text{up}}(H)]dH + \int_{0}^{H_c} [R_{\text{up}}(H) - R_{\text{down}}(H)]dH,$$

(1)

where $H_c \approx 1200$ Oe is the larger coercive force in the two FM$s$, and $R_{\text{up}}(H) (R_{\text{down}}(H))$ is magnetoresistance in increasing (decreasing) magnetic field. The calculated SIMR is shown in Fig. 3(a) as a function of the gate voltage. For comparison, the two-terminal resistance of the device measured at $H = 0$ is plotted in Fig. 3(b). Positive correlation between SIMR and resistance is clearly seen, indicating that the inverse spin valve effect is related to small device resistance.

4. Discussion
We now discuss the origin of the inverse spin valve effect. First, we remark that the effect is not originated from the extrinsic magnetoresistance such as anisotropic magnetoresistance or Hall effect, because these MRs depend only on magnetization alignment in ferromagnetic electrodes and are not affected by gate voltage or resistance. There are some candidates which reverse the spin valve effect; these are

1. the Rashba spin-orbit scattering [17],
2. the quantum interference [18] such as resonant states through Coulomb blockade [2], Fabri-Pérot interference [3] or wave vector matching [4], and
3. the ferromagnet-oxide interface [1, 10].

Using FM/graphitic material/FM junctions, several groups have reported the inverse spin valve
Figure 4. Calculated SIMR (solid line) and $G_{AP}$ (dashed line) as a function of transmission probability parameter

Effect due to not only gate voltage [2–4, 7, 10], but also temperature [1] and source-drain bias [9] and their results are attributed to the above mentioned origins.

Our results can be explained by the quantum interference, assuming that the transmission probabilities at the two interfaces increase with the gate voltage. In the Fabri-Pérot interference model [3], the transmission probability for the majority spin (+) and minority spin (−) of the left(s) and right (r) contacts is defined with

$$T_{sr} = T_L T_R \sum_{n=0}^{N} [\left(1 - T_L^n\right)\left(1 - T_R^n\right)]^{n/2} e^{2i\pi n\delta},$$

(2)

where, following Refs. [2, 3], $T_{L(R)}^{n}$ is spin-dependent transmission probabilities at the left (right) contacts. Here, $T_{L(R)}^{n} = T_{L(R)}(1 + sP)$, $s = \pm 1$ and spin polarization $P$ is assumed to be 0.23 [19]. The phase $\delta$ acquired by an electron at the round trip between two electrodes is fixed to be zero, and the repetition number $N$ coherently reflected at the electrodes is set to be 2. We define the transmission probability parameter $t$ with $T_L = 0.75t$ and $T_R = 0.25t$ and plot the SIMR $\equiv [(T_{++} + T_{--})^{-1} - (T_{++} + T_{--})^{-1}] / [(T_{++} + T_{--})^{-1}]$ and $R_{AP} \equiv (T_{++} + T_{--})^{-1}$ as a function of $t$ in Fig. 4. The SIMR is positive at small $t$, but turns to negative with increasing $t$. $R_{AP}$ is also a simply decreasing functions of $t$. Even though this calculation is oversimplified because the $V_g$ dependence of $\delta$ and the number of conductance channels are neglected, the result is consistent with the correlation between the SIMR and the resistance shown in Fig. 3.

We emphasize that the quantum interference indicates the electrons and spins transport coherently in MLG, with $l_e$ and $\lambda_N$ substantially longer than the electrode spacing, 300 nm in the present sample. The mean free path of 300 nm implies the mobility of 12,000 cm$^2$·V$^{-1}$·s$^{-1}$, which is easily attained in MLG at low temperatures. The long spin relaxation length is also consistent with our previous report [11] which shows $\lambda_N$ much larger than 8.4 $\mu$m.

Finally, we estimate the intrinsic spin coherence length in MLG. Without magnetic impurities, the spin-orbit scattering is responsible for the spin relaxation. The intrinsic spin-orbit scattering time is given by $\tau_{so} = (\alpha Z)^{-4}\tau_e$ [20], where $\tau_e$ is the elastic scattering time, $\alpha = 1/137$ is the fine structure constant, and $Z$ is the atomic number. The spin relaxation length in MLG is estimated to be $\lambda_N = (\alpha Z)^{-2}l_e \sim 500l_e$. Substituting the typical value $l_e = 100$ nm, we obtain macroscopically long spin relaxation length $\lambda_N = 50$ $\mu$m. Thus, the light atomic mass and the large mobility make MLG a promising material for spintronics and related applications.
5. Conclusion
In conclusion, we succeeded in controlling the spin conduction in MLG by the electric field, which was confirmed by the local measurement. The inverse spin valve effect was observed by applying the large gate voltage. The results suggest the quantum coherence in MLG, which signifies that the mean free path and the spin relaxation length in MLG are longer than electrode spacing, 300 nm. The gate tuning of MR opens the new possibility of graphite-based spintronic devices and MLG is a promising material for its realization.

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