Large local Hall effect in pin-hole dominated multigraphene spin-valves

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Abstract
We report local and non-local measurements in pin-hole dominated mesoscopic multigraphene spin-valves. Local spin-valve measurements show spurious switching behavior in resistance during magnetic field sweeping similar to the signal observed due to spin injection into multigraphene. The switching behavior has been explained in terms of a local Hall effect due to a thickness irregularity of the tunnel barrier. The local Hall effect appears due to a large local magnetostatic field produced near the roughness in the AlO$_x$ tunnel barrier. In our samples the resistance change due to the local Hall effect remains negligibly small above 75 K. A strong local Hall effect might hinder spin injection into multigraphene, resulting in no spin signal in non-local measurements.

(Some figures may appear in colour only in the online journal)

1. Introduction
Graphene and few-layer graphene has been shown to be an ideal material for spintronics devices because of its suppressed hyperfine interaction and weak spin–orbit coupling [1]. Gate tunable conductivity along with low carrier density ($n < 10^{12}$ cm$^{-2}$) and high mobility ($\mu \sim 10^4$ cm$^2$ V$^{-1}$ s$^{-1}$) give the opportunity to fabricate ultra-fast spintronic devices with this material [2]. Efficient spin-polarized carrier injection into graphene is one of the essential requirements for such devices. Significant improvement has been achieved in this direction through successful demonstration of spin injection into single- and few-layer graphene (also called multigraphene) at room temperature by different groups [3–10]. Long spin diffusion length $\lambda_s \sim 2 \mu$m and spin life-time $\tau_{sp} \sim 50–200$ ps have been reported in single-layer graphene through spin injection experiments [3]. Much longer spin life-time $\tau_{sp} \sim 2$ ns has been observed in bilayer graphene [11, 12]. Few-layer graphene is now believed to be a more appropriate candidate for spin injection compared to single-layer graphene because of the screening of scattering potentials from the substrate in the former [12–14]. However, experimental values of $\lambda_s$ and $\tau_{sp}$ are still an order of magnitude shorter than what is expected theoretically [1]. Therefore, an efficient engineering of spin-valve devices is required to realize the full potential of multigraphene spintronics devices.

Spin-valve devices for spin injection show high to low resistance switching, which depends on the relative magnetization of the two ferromagnetic electrodes. Spin-polarized carriers are injected from one ferromagnet (injector) and the probability of these carriers to be collected at the other ferromagnet (detector) depends on the magnetization of the latter. Therefore, the quality of interface between the ferromagnet and graphene may significantly affect the spin-polarized carrier injection into graphene. A proper understanding of the micromagnetic phenomena occurring at the interface is essential for the optimization of such devices. In order to avoid conventional Schottky barrier contact due to the large conductivity mismatch between graphene and the metallic ferromagnet, a tunneling approach has been suggested [15, 16]. However, conventional tunnel barriers suffer from complications such as pin-holes and nonuniform barrier thickness. These defects in the barrier give additional features by contributions from anisotropic magnetoresistance (AMR) and local Hall effect that obscure the spin injection signal [17]. In particular, stray magnetostatic fields produced at the interface due to the ferromagnet on top of it may have
significant impact. Recently, spin precession and inverted Hanle effect have been reported in Si spin injection devices; these arise from local magnetostatic fields at tunnel barrier roughness [18]. In this work, we demonstrate large local Hall effects due to stray magnetostatic fields at the tunnel barrier roughness in multigraphene spin-valves. We observed resistance switching behavior due to this local Hall effect. We found that spin injection is strongly hindered by the presence of these stray magnetostatic fields. No resistance switching was observed in non-local measurements due to this.

2. Fabrication and experimental details

Multigraphene samples were prepared on Si/SiN \( \text{N}_4 \) substrate by rubbing a small flake of highly oriented pyrolytic graphite (HOPG) from Advanced Ceramics (grade ZYA, rocking curve 0.4°). The thicknesses of the multigraphene flakes were measured using an atomic force microscope (AFM). For device fabrication ~30 nm thick and ~10 \( \mu \)m long multigraphene samples were chosen. Spin-valve devices were fabricated by the conventional electron beam lithography method. Thermally evaporated Al on top of multigraphene was oxidized to form an \( \text{AlO}_x \) tunnel barrier. The thickness of the \( \text{AlO}_x \) tunnel barrier was found to be ~2 nm from AFM measurements. For spin-polarized carrier injection into multigraphene ~50 nm thick Co was thermally evaporated on a pre-patterned structure of PMMA prepared by electron beam lithography. Ferromagnetic Co lines of different widths (as shown in figure 1) were patterned using this technique. Different widths of Co were used to ensure different coercive fields between electrodes. The distance between two inner Co electrodes (l) was varied from 1 to 3 \( \mu \)m for different spin-valve devices. A thin layer of Pt was immediately evaporated on top of Co to prevent it from oxidation. The Co lines were further contacted to larger contact pads through gold lines. Magnetoresistance measurements were made in a closed cycle refrigerator using an AC resistance bridge. For spin-valve measurements magnetic field was applied in the plane of the film along the long axis of the Co line using an electromagnet with rotation option.

3. Results and discussion

The temperature dependence of the resistance of the spin-valve device showed a semiconducting behavior as typically observed for multigraphene samples reported elsewhere [19, 20]. Current–voltage (I–V) characteristics were measured on as-fabricated samples at room temperature and 15 K. Spin-valve devices showed slightly nonlinear I–V with a few k\( \Omega \) resistance due to the presence of an \( \text{AlO}_x \) tunnel barrier. However, with repeated I–V measurement at maximum current up to 10 \( \mu \)A the devices showed linear I–V with resistance decreasing to ~435 \( \Omega \) at 15 K. The rapid decrease of resistance with linear I–V can be understood by considering formation of pin-holes during the measurement process. The resistance of the spin-valves was measured with an AC resistance bridge with 1.5 \( \mu \)A current limit range. Figure 2 shows the field dependence of the resistance of one of the spin-valve devices measured at 15 K. The measurement configuration called the local configuration is shown in the inset of figure 2. Magnetic field (B) is applied along the long axis of the Co line. Resistance switching can be seen at a magnetic field \( B \sim 0.1 \, \text{T} \). The resistance switches from a higher resistance to a lower resistance while the field is swept from +0.5 to −0.5 T. The reverse happens while the field is swept from −0.5 to +0.5 T. The direction of field sweeping is shown by arrows in the figure.

Figure 3 shows a similar resistance switching behavior for a three-terminal local configuration as shown in the inset. Our resistance switching behavior is significantly different from the resistance switching seen in local measurements in graphene spin injection devices reported in the literature [3, 11]. In spin injection devices step-like switching is seen for parallel and antiparallel orientation of the ferromagnetic electrodes. A high resistance state is achieved for an
Figure 3. Local spin-valve measurements in three-terminal local configuration as shown in inset. Magnetic field is applied along the long axis of the Co electrodes.

Figure 4. (a) AFM image of multigraphene with AlO$_x$ on top of it. (b) Roughness profile along the line in (a).

antiparallel configuration of the inner two electrodes and a low resistance for a parallel configuration. Our switching behavior is quite unusual and can be misunderstood as a signature of the spin injection. The observed switching behavior can be understood by considering the presence of different stray magnetostatic fields in the spin-valve device. Magnetostatic fields in a spin-valve device appear mainly from two sources: (1) fringe magnetostatic fields at the edges of the ferromagnetic electrodes [21] and (2) magnetostatic fields near the multigraphene and ferromagnet (Co) interface due to finite roughness of the tunnel barrier [18]. With the magnetic field applied along the long axis of the Co electrode, fringe fields from the edges can be ignored in our spin-valves, as the short edges of the Co electrode lie far from the multigraphene (see figure 1). Most of the magnetostatic fields in our device mainly originate from the AlO$_x$ roughness.

Figure 5. (a) The out of plane component of magnetic field $B_z$ calculated from equation (1) for two different roughness amplitudes $h = 1$ and 2 nm. Here $z = 2$ nm and $\lambda \sim 20$ nm. (b) Sketch of magnetostatic field from a sinusoidal roughness profile for two different directions of magnetization of the Co electrode. Magnetic poles developed at the interface are shown by arrows.

along the $x$ axis the magnetostatic field generated from a ferromagnet with magnetization pointing along the $x$ direction can be written as [18, 22]

$$B_x(x, z) = \mu_0 M_s \frac{h}{2} \sum_{n=1}^{\infty} q_n F(q_n) e^{-q_n z} \sin(q_n x - \pi/2)$$

$$B_y(x, z) = 0$$

$$B_z(x, z) = \mu_0 M_s \frac{h}{2} \sum_{n=1}^{\infty} q_n F(q_n) e^{-q_n z} \cos(q_n x - \pi/2)$$

where $q_n = 2\pi n/\lambda$ and

$$F(q_n) = \frac{\sin(q_n \lambda/4) \sinh(q_n h/2)}{(q_n \lambda/4) \sinh(q_n h/2)}.$$  

Here $h$ is the peak-to-peak roughness height and $\lambda$ the period of the roughness profile of the barrier. To get a detailed idea about the roughness of the AlO$_x$ tunnel barrier we performed AFM measurements of our spin valve, as shown in figure 4(a). The roughness profile of AlO$_x$ on top of multigraphene along the marked line in figure 4(a) is shown in figure 4(b). Note that in our spin-valves multigraphene is completely covered with AlO$_x$ before patterning the Co electrodes. Clearly, the roughness amplitude in the AlO$_x$ layer is $h \sim 1$ nm. However, considering the linear $I$–$V$ characteristics and the possibility
to have pin-holes, we can assume a maximum roughness amplitude $h \sim 2$ nm, i.e. of the order of the thickness of the AlO$_x$ barrier. For ferromagnetic Co with $\mu_0 M_s = 1.82$ T, and assuming $\lambda \sim 20$ nm, the out of plane component of magnetic field ($B_z$) calculated using equation (1) at $z = 2$ nm is shown in figure 5(a) for $h = 1$ and 2 nm, respectively. The local magnetostatic fields can be as large as $\sim 0.45$ T. A pictorial representation of the stray magnetostatic fields for a sinusoidal roughness of the tunnel barrier is shown in figure 5(b) with magnetization pointing in two opposite directions. Therefore, when an external field $\sim 0.1$ T is applied to saturate the Co electrode, the multigraphene with a tunnel barrier roughness $\sim 2$ nm can experience much higher field of the order of 0.45 T in local areas. We believe these stray magnetostatic fields will provide an additional voltage contribution $V_{\text{inh}}$ to the measured longitudinal voltage $V_{\text{xx}}$. The value of $V_{\text{inh}}$ will depend on the actual distribution of the stray magnetostatic fields, and therefore on the roughness. However, in a more qualitative way one can say that in a mesoscopic device with irregular geometry the voltage between any two points can be written as a linear combination of longitudinal ($V_{\text{xx}}$) and Hall ($V_{\text{xy}}$) voltages. Therefore, one can write $V_{\text{inh}} = aV_{\text{xx}} + bV_{\text{xy}}$, where $a$ and $b$ are sample dependent constants. When the magnetization of the Co electrode changes direction, the perpendicular component of the magnetostatic field $B_z$ changes sign, resulting in a change in the sign of the Hall voltage $V_{\text{xy}}$. Therefore, the measured voltage $V_{\text{meas}}$ for $B_z$ pointing upward, can be written as $V_{\text{meas}} = V_{\text{xx}} + V_{\text{inh}} = (a + 1)V_{\text{xx}} + bV_{\text{xy}}$, and in this case one gets a high resistance state. Similarly, for $B_z$ pointing downwards one can write $V_{\text{meas}} = (a + 1)V_{\text{xx}} - bV_{\text{xy}}$ and thus a low resistance state is achieved. The switching from the high resistance to low resistance state occurs at a magnetic field when magnetization of the Co electrode changes, i.e. at the coercive field of the inner Co electrode. Although one expects two resistance switchings corresponding to the magnetization switching of the two inner Co electrodes, we found only one resistance switching, probably because the narrower Co electrode produces too small a resistance change to be observable within our experimental resolution. We also measured our spin-valves at different temperatures and we found that the resistance change produced by this local Hall effect becomes too small and in the range of experimental noise above 75 K. The field dependence of resistance measured at $T = 50$, 60, 75 and 100 K in the local configuration (see figure 2) is shown in figure 6. The temperature dependence can be understood by considering the expression $V_{\text{meas}} = (a + 1)V_{\text{xx}} \pm bV_{\text{xy}}$. A significant effect due to the local Hall effect can be observed only if $V_{\text{xy}}$ is large. In multigraphene samples $V_{\text{xy}}$ (or $R_{\text{xy}}$) is found to increase with decreasing temperature. Therefore, below $\sim 75$ K, $V_{\text{xy}}$ becomes large enough to produce a measurable resistance change.

Another possible source of switching in resistance in spin-valves can be anisotropic magnetoresistance (AMR) of the Co electrodes [23]. However, we could not find any sharp resistance switching in spin-valves with Co electrodes without any AlO$_x$ tunnel barrier [24]. Therefore, we believe the sharp resistance switching in our spin-valves is mainly due to the local Hall effect.

A clear signature of spin injection into multigraphene is usually seen in non-local measurements in a configuration as shown in the inset of figure 7. In this configuration, the electrical charge current path is completely separated from the spin current path so that only signals due to spin current can be observed. In our devices, we could not find any switching in resistance in a non-local measurement made at 15 K with voltage pads placed 1.5 $\mu$m away from the current path. We

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**Figure 6.** Local spin-valve measurements at (a) $T = 50$ K, (b) $T = 60$ K, (c) $T = 75$ K and (d) $T = 100$ K in the configuration shown in the inset of figure 2. The absence of resistance switching above 75 K shows a reduced effect of the local Hall effect.
believe the local Hall effect due to tunnel barrier roughness significantly suppresses spin injection into multigraphene. Although there might be additional reasons for the absence of non-local signal in our spin-valves, we believe that local magnetostatic fields at the tunnel barrier roughness definitely play a role in spin injection. Therefore, for a clear observation of resistance changes due to spin injection into multigraphene, full coverage of the spin-transport channel with a smooth tunnel barrier is essential.

4. Conclusions

In summary, we have fabricated submicron spin-valve devices with few-layer graphene of different thicknesses. Our spin-valves showed resistance switching behavior at low temperatures in the local measurement configuration. However, in the non-local configuration we could not see any switching, indicating reduced spin injection. The switching behavior in local measurement can be qualitatively understood in terms of a large local Hall effect. The local Hall effect appears due to magnetostatic fields present in the device. We estimated that for a roughness $\sim 2$ nm the local magnetostatic fields can be as large as 0.45 T. The local Hall effect can result in spurious resistance switchings, which can be confused with resistance switching due to spin injection into spin-valve systems. Pin-hole free smooth barriers along with narrow ferromagnetic electrodes are prerequisite for spin injection into multigraphene.

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