

Local long-distance spin transport in single layer graphene spin filter

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(Presented 8 November 2017; received 2 October 2017; accepted 3 November 2017; published online 21 December 2017)

Spin transport through single layer graphene grown by chemical vapor deposition has been measured in the local measurement configuration using aluminum oxide tunnel barriers and Co electrodes. Unlike the case of non-local measurements we report positive magnetoresistance for graphene channels as long as 10 microns at room temperature. Our analysis shows that the spin diffusion length in graphene is 14 ± 4 microns at room temperature, bringing us a step closer to spintronic applications based on graphene spin filters. © 2017 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>). <https://doi.org/10.1063/1.5007661>

Spintronics devices compatible with flexible electronics is a requirement for novel technologies which combine logic and memory on the same chip.¹⁻³ This would require a flexible interlayer and graphene is a perfect material for such an application. Due to its low atomic number carbon based interlayers, such as carbon nanotubes have been studied as an interlayer in spin valve devices,⁴ but due to their difficult fabrication routines they have not been considered for real applications. Graphene, on the other hand would be more suitable since its 2 dimensional crystal structure can be obtained in wafer scale and they are much more reproducible compared to nanotubes.⁵

Owing to its extraordinary electronic and magnetic properties graphene has become the topic of research for condensed matter physics in more than ten years.⁶ Due to its high mobility and long mean free path the spin transport in graphene should be realized over long distances. The spin valve geometries used so far for graphene based devices has measured the spin signals in the non-local geometries and the longest measured to date spin diffusion length was $16 \mu\text{m}$,⁷ even though the theoretical calculations suggest it can be in the order of $100 \mu\text{m}$.⁸ There are several factors affecting the spin transport through graphene. First, the free electron density difference at the contact areas create a huge potential causing spin flip probability to increase and thereby prohibiting the flow of spins through graphene.^{1,2,9} This is a common problem with several spin injection cases with semiconductors, but this has been solved to a large extent through the use of tunnel barriers at the contact regions.¹⁰ Second, the quality of graphene layers differ a lot based on the method used to prepare the samples. The initial works were on single layer graphene on SiC substrates, where the purity of graphene and single crystal nature of it could be an advantage^{1,3,11} because graphene layers grown by chemical vapor deposition (CVD) is polycrystalline and may have much more defects and wrinkles.¹² Later Kamalakar *et al.* have demonstrated that CVD graphene layers can also have very long spin diffusion length.⁷ These works realize the spin transport measurement in the non-local geometry where the drift current is separate from the voltage leads. Here we report spin diffusion lengths of 14 micrometer at room temperature through CVD grown single layer graphene in the local measurement configuration.

Single layer graphene layers have been grown on 25 micron thick high purity Cu foils. Before loading the copper foils in the CVD furnace we clean them in acetone and isopropanol using ultrasonic

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agitation and blow dry. Then we flattened the copper foils using a roller press, which produces a mirror like finish. To grow graphene we load a 20×100 mm long piece of copper foil in a 22mm inner diameter quartz tube and flow 200 sccm Ar (6N purity), and 50 sccm H_2 (5N5 purity) and raise the temperature to 1000°C and keep this gas flow steady at 1000°C for 1 hour. Then the graphene growth is done with 10 sccm CH_4 (5N5 purity), 5 sccm Ar and 5 sccm H_2 for 10 minutes. Then CH_4 gas is closed and 200 sccm Ar and 50 sccm H_2 is flown during a slow cool down which takes approximately 2 hours. The graphene layers on copper foil were separated by dissolving copper in a 1M ferric chloride bath and they were transferred to deionized water bath in a watch glass. Few drops of dilute HCl are used in this bath to remove any possible iron contamination from the ferric chloride bath. Then the graphene layers have been washed in new deionized water baths and then subsequently transferred to a Si/SiO_x wafer piece using standard transfer methods. Initially there is some water under the graphene layer, and we let this water to dry slowly in a refrigerator, which we found gives better adhesion on silicon oxide.

Spin filters have been designed and fabricated using standard microlithography, sputter deposition and plasma etching techniques. Thin films and plasma etching processes were performed in a high vacuum chamber with a base pressure of 1×10^{-7} Torr. Figure 1 shows a schematic of steps of the spin valve fabrication process. Ferromagnetic electrode shapes $1 \times 20 \mu\text{m}$ and $3 \times 20 \mu\text{m}$ have been defined to have different coercivities for magneto transport measurements. Standard ultraviolet microlithography techniques were used to fabricate graphene spin filters. First, the shapes of Co electrodes were defined on a blanket transferred single layer graphene on Si/SiO_x substrate, then approximately 1 nm thick AlO_x tunnel barrier has been deposited by sputtering 0.2nm Al and then oxidizing in the vacuum chamber in ambient oxygen at 1×10^{-2} Torr partial pressure of pure oxygen, and this process has been repeated 4 times to get ~ 1 nm thick AlO_x tunnel barrier. This method has been proven to give almost pinhole free AlO_x tunnel barriers.^{11,13} Without breaking vacuum 10 nm thick Co was deposited by sputtering. Co target has been cleaned by pre-sputtering before deposition on AlO_x tunnel barrier. Then excess areas on graphene layers were defined by lithography and removed by oxygen plasma. Top contact layers Ta (3nm)/Cu (50nm) were deposited afterwards. Deposition and lift-off techniques were used throughout the microfabrication steps.

Magneto transport measurements were performed on the fabricated samples at room temperature using four-terminal wiring as shown in Figure 2, where 10 μA excitation current is used during measurements. Figure 3 shows a typical Magnetoresistance (MR) curve for a 10 micron graphene channel at room temperature. This result is interesting from the point of view that the MR

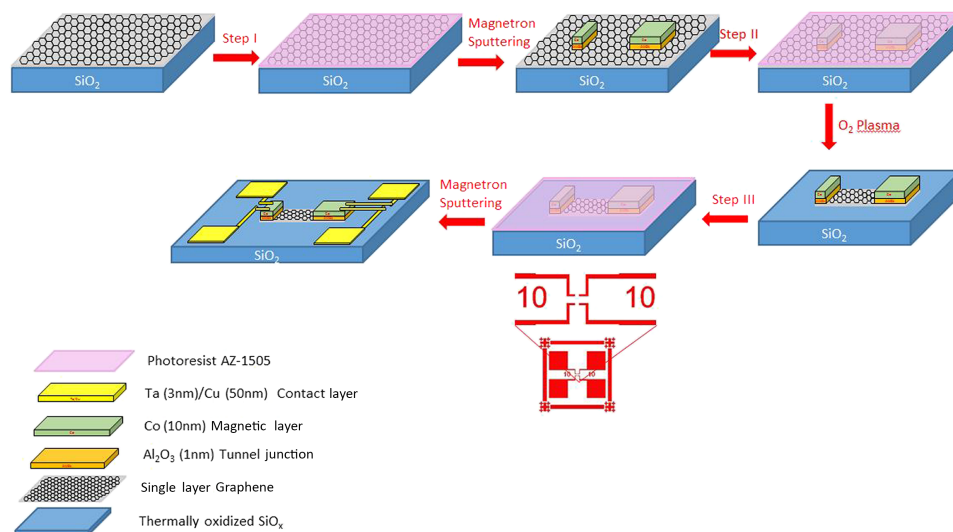


FIG. 1. Micro fabrication steps of graphene spin filters.

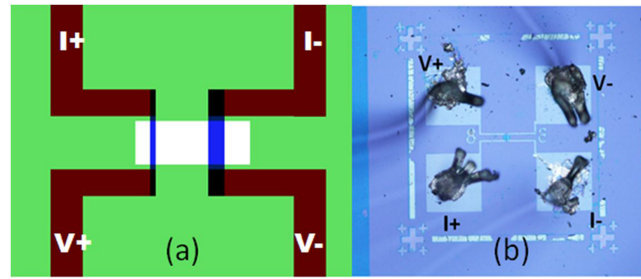


FIG. 2. (a) Central part of spin filter design and for terminals labelled (b) Actual graphene spin filter device.

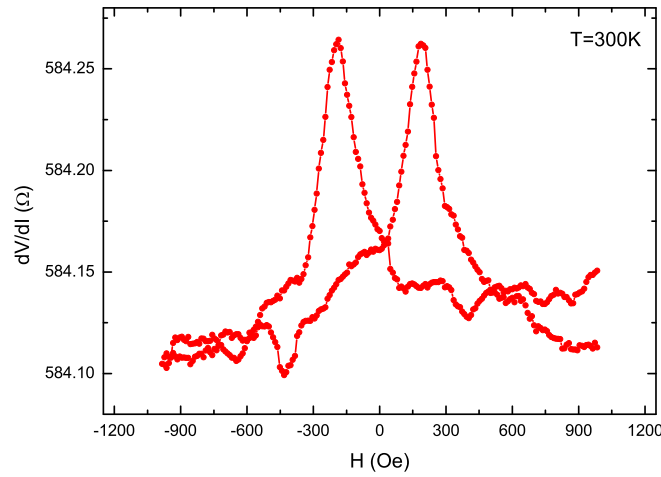


FIG. 3. Magnetoresistance curve of a 10 micron channel single layer graphene spin valve at room temperature.

measurements with graphene spin filters measured in the non-local geometry all show negative MR¹⁴ while our results clearly and reproducibly show positive MR in the local measurement configuration in agreement with the results of Dlubak et al.,⁸ where they demonstrate spin transport through 2 micron channels of graphene on single crystal silicon carbide substrates. Subsequent works simply utilized a non-local spin injection and detection scheme where pure spin currents can give much

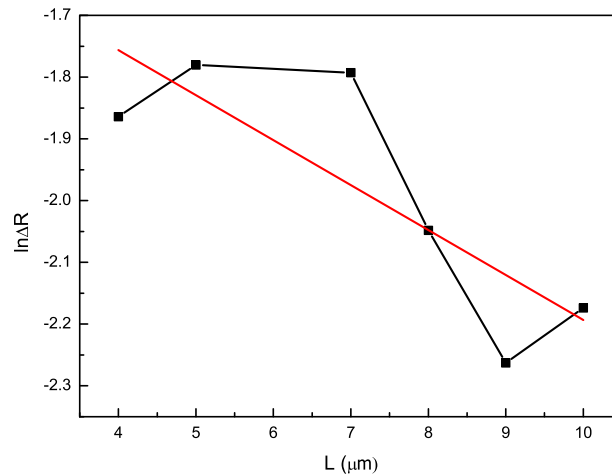


FIG. 4. Natural logarithm of magnetoresistance vs. graphene channel length at room temperature.

better results, but this measurement geometry is not a real device terminal geometry, which utilizes terminals for both current injection and detection across the spacer layer. Spin diffusion length can be estimated from $\Delta R \propto \exp(-L/l_{sf})$ where l_{sf} is the spin diffusion length.⁷ We have fabricated spin filters with 4,5,7,8,9 and 10 micron single layer graphene channels and plotted their $\ln \Delta R$ vs L as shown in Figure 4. Our results vary a lot and do not directly follow an exponential decay, which we think is due to the inconsistencies of the tunnel barrier. Moreover, spurious signals can arise due to non-uniform current effects both in the graphene layer and also the interfaces with the tunnel barrier. According to the linear fit of these data points we found that the spin diffusion length in graphene is $14 \pm 4 \mu\text{m}$, which is in close agreement for the measurements reported in the work of Kamalakar et al.⁷

In conclusion, we have demonstrated spin transport through 10 micron long CVD grown single layer graphene at room temperature. The magnetoresistance has a positive sign unlike in the case of non-local measurements. Our results bring us a step closer to fully functional spin filters based on graphene which can be integrated in combined logic and memory chips in future flexible electronic devices.

ACKNOWLEDGMENTS

This work was supported by Turkish Scientific and Technological Research Council (TUBITAK) under project number 113F161.

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