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TRANSVERSE SPIN TRANSPORT IN GRAPHENE

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In this paper we report transverse spin transport properties of graphene in a device, where for the first time a mono-atomically thin atomic fabric was sandwiched between magnetic thin films. We found that a single layer graphene flake was sufficient to break the exchange coupling between magnetic films and also to enhance the magnetoresistance effect.

Keywords: Graphene; magnetoresistance; permalloy.

1. Introduction

Graphene continues to attract massive research interest as a novel and exciting research material. Outstanding electronic properties and excellent compatibility with existing semiconductor technology makes it a real contender for replacing silicon. These perspectives are particularly promising in a new field of spintronics. The crystal structure of grapheme represents a mono atomically thin 2-D film of Carbon atoms arranged laterally in a honeycomb benzene ring like structure. Graphene is a zero gap semiconductor, where the charge carriers have a linear dispersion relation near the Dirac point. The ambipolar electric field effect in graphene makes it possible to control the charge carriers with concentrations of up to 10^{13} cm⁻² (Refs. 1-3) and room temperature carrier mobilities of ~ 10^4 cm²/Vs are routinely observed.^{4,5} These mobilities were found to be weakly temperature dependent, which means that if impurity scattering was reduced, ~200,000 cm²/Vs mobilities could be achieved. Transport in this system is ballistic and carriers can travel submicrometer distances^{6,7,8} (~300 nm) without scattering. In addition, the charge carriers in graphene were found to behave like a 2-D gas of massless Dirac fermions,⁴ and the quantum Hall effect was observed at room

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temperature.⁹ Collectively these factors not only make graphene a novel material for electronics but also for spintronics research. The opportunity to control the charge carriers in graphene along with their spin by an applied gate voltage adds to its advantages for applications in spintronics.

Graphene has been shown to support spin polarized conduction in the plane of the film $(CIP - conduction in plane)^{10-12}$ when spin polarized current was injected into graphene with ferromagnetic electrodes. However, no study has been done prior to this to employ graphene in devices where the conduction is perpendicular or transverse to plane (CPP). In this paper we describe the study of transverse spin transport properties in graphene by using it as a non-magnetic spacer layer sandwiched between metallic magnetic thin films.

2. Device Fabrication

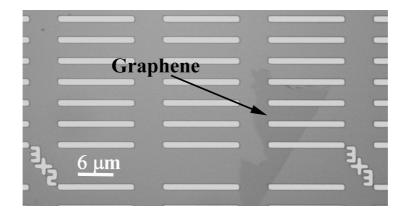


Fig. 1. An optical image showing the repeated bottom electron array structure designed by e-beam lithography. Graphite flakes are peeled and then deposited on this structure.

Oxidized (100 nm SiO₂) n-doped Si substrates were patterned using e-beam lithography with a repeated grid like structure consisting of ~1 μ m wide lines ~20 μ m in length. Each block of grid lines was supplied by numbered reference marks for locating and alignment purposes. The pattern is made sufficiently dense, covering a large portion of the substrate to increase the probability to deposit some of graphene flakes above an element in the matrix. This patterned mask was used to prepare a bottom electrode array by sputtering of permalloy, Py (Ni₈₀Fe₂₀). Figure 1 shows a part of this bottom electrode grid structure on the substrate.

Graphene is obtained by repeated peeling of $\sim 1 \text{ mm}^2$ flakes of natural graphite. The peeled flakes are then transferred onto the substrate. Such a technique¹ results in high quality crystallites up to $\sim 1000 \,\mu\text{m}^2$ in size. After deposition the substrate is inspected

under an optical microscope to detect weak but visible changes in optical contrast which allows location of monolayer graphene flakes.¹³ One of such a single layer flakes is shown in Fig. 1. Single layer graphene demonstrates a clear signature in Raman spectroscopy.^{14,15} Therefore we verified some of the flakes with this method.

Care is taken to remove any possible contamination before depositing the films. The substrate is cleaned in Ar plasma in a vacuum chamber and immediately after that graphene flakes are deposited keeping the exposure time of the substrate to air at a minimum.

After locating a graphene flake another step of e-beam lithography is carried out to design the top electrodes. This lithography step also involves producing contacts to the upper and lower electrodes for measurement purposes. For comparison we also always fabricated a reference all metal Py device without graphene.

3. Measurements and Results

Simple cross electrode resistance measurements were used to study transverse electric transport as shown in Fig. 2(a). The resistance was measured as a function of magnetic field. The magnetic field was aligned parallel to the plane of the substrate and along the length of the bottom electrode.

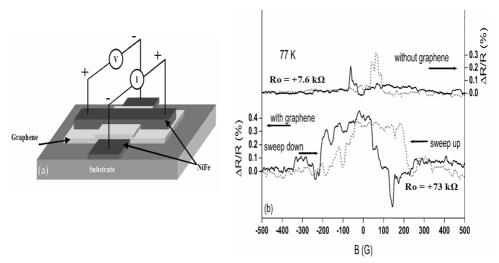


Fig. 2. (a) Schematic diagram of the transverse spin transport device and the configuration of the electrodes used for all measurements. The graphene flake is sandwiched between the layers of Py electrodes. (b)~Magnetoresistance (MR) response of the device using only plain Py electrodes with and without the graphene flake at 77 K.

Due to the nature of preparation technique the bottom Py electrode is subjected to interaction with air, which leads to creation of an oxide layer on its surface. This strongly affects the experimental behavior of the magnetoresistance (MR). Figure 2(b) demonstrates typical behavior of MR for devices with plain unprotected bottom Py electrodes. A thin oxide layer in this case increased the contact resistance to the flake. It was however not sufficiently continuous to magnetically uncouple the permalloy electrodes when the flake was absent as seen in Fig. 2(b). Thus, we only observed spikes in the resistive response which are similar to those observed on single layer films and are attributed to AMR (Anisotropic Magnetoresistance) or domain wall scattering as the film switches. However, with the flake there was a stable high resistance state at zero magnetic field indicating non-parallel magnetization states for upper and lower magnetic electrodes. Without the graphene flake the films appear to be sufficiently coupled to prevent magnetic re-ordering at zero field since the resistance is the same as that in the high field state where the magnetizations would be aligned parallel. In contrast with the graphene present the zero field resistance is significantly different from the parallel case indicating the presence of a GMR (Giant Magnetoresistance) effect. Efficient magnetic coupling in plain devices may possibly be related to pin-holes in the oxide layer. Surprisingly, our results indicate that the one atomic layer thick graphene flake is sufficient to reduce this coupling drastically. Studying the current voltage characteristics we observe tunnel like behavior in this case, we believe this is due to the oxide layer on the surface of bottom the Py film.

To overcome the problem of the oxide layer the bottom permalloy contacts were capped with a Au layer approximately 2 nm thick. For capped electrodes we also find qualitatively different behavior of devices with graphene and without graphene. For a measurement configuration shown in Fig. 2(a) a negative voltage was obtained from the output terminals "V" in the case of the device without graphene. As can be seen from the left sketch of Fig. 3(a) the negative sign is expected for a dominant CIP resistive component when the interface layer is not important and the current distribution could be modeled by a homogenous cross with a top square in the middle. For an opposite case of an insulating interface layer, top and bottom electrodes become equipotential resulting into a positive sign for the measured voltage (Fig. 3(a) right). The positive voltage is observed for the device with the graphene flake. This indicates that the resistive component is now more dominated by the current flowing perpendicular to the device plane (CPP geometry). For devices with capped bottom electrode the I-V curves were found to be linear in both cases with and without graphene. Consequently, tunneling is thought to play no significant role.

Fig. 3(b) shows the MR ratio response derived from the resistance measurements. These are absolute values taken ignoring the sign difference between the devices. A MR response is present in both cases. However, in the device with graphene, the MR response was larger by a factor of four. In a conventional spin valve it would be expected that at very high magnetic fields there would be parallel magnetization between the top and bottom layer and hence lower resistance. We observed that a significant anisotropic MR

is present in our devices. When the direction of the applied magnetic field was rotated orthogonal to the direction used in measurements shown in Fig. 3(b), the MR of the device with graphene dropped to 2%. It is clear from this that a significant negative MR component is present, which depends on the angle of the applied field and behaves similarly to the results obtained by Suezawa and Gondo for Ni/Co cross junctions.¹⁶

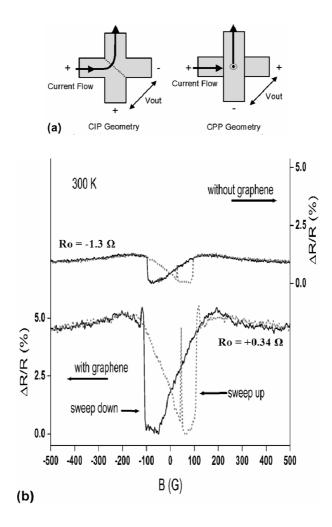


Fig. 3. (a) A schematic illustration of the observed reversal of the output potential "V" in measurement configuration of Fig 2(a). Left and right sketch correspond to the CIP and CPP geometries respectively. (b) A MR response derived from the absolute values of the resistance measured for a typical device with the Au capping layer. The top curve corresponds to the reference device without graphene. A spin valve response is seen for both devices. In the case of the device with graphene present the response is larger by a factor of four.

4. Conclusion

Our transverse spin transport study indicates that the electric current is being channeled through graphene, which is evident from the change in sign of the measured voltage on the cross with and without graphene. This channeling is probably taking place in the area of defects in the graphene flake sandwiched between the top and bottom magnetic layer.

We conclude that for Au capped bottom electrodes in graphene devices the increase in the MR is due to the CPP conduction becoming more important unlike the reference device without the graphene flake where the CIP component dominates. In devices without capping layer we find that a single layer flake of graphene significantly weakens the exchange coupling between two ferromagnetic layers suppressing the parallel magnetization state at low magnetic fields.

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References

- 1. K. S. Novoselov et al., Science 306, 666 (2004).
- 2. K. S. Novoselov et al., Proc. Natl. Acad. Sci.USA 102, 10451 (2005).
- 3. A. K. Geim and K. S. Novoselov, Nature Mater. 6, 183-191 (2007).
- 4. K. S. Novoselov et al., Nature 438, 197 (2005).
- 5. Y. Zhang et al., Nature 438, 201 (2005).
- 6. S. Morozov et al., Phys. Rev. Lett. 100, 016602 (2008).
- 7. K. Bolotin et al., Solid State Commun. 146, 351 (2008).
- 8. X. Du et al., Nature Nanotech. 3, 491 (2008).
- 9. K. S. Novoselov et al., Science 315, 1379 (2007).
- 10. E. W. Hill et al., IEEE Trans. Magn. 42, 2694 (2006).
- 11. N. Tombros et al., Nature 448, 571 (2007).
- 12. S. Cho et al., Appl. Phys. Lett. 91, 063124 (2007).
- 13. P. Blake et al., Appl. Phys. Lett. 91, (2007).
- 14. A. C. Ferrari et al., Phys. Rev. Lett. 97, 187401 (2006).
- 15. D. Graf et al., Nano Lett. 7, 238 (2007).
- 16. Y. Suezawa and Y. Gondo, J. Magn. Magn. Mater. 126, 524 (1993).