

Graphene in Multilayered CPP Spin Valves

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In this paper, we have sandwiched a single-atom-thick sheet of graphene, a 2-D allotrope of carbon that is exciting tremendous research interest, between two ferromagnetic electrodes. Graphene has already been shown to support spin polarized conduction, when spin polarized electrons were injected into graphene sheets along the plane using magnetic electrodes. Here, we study spin polarized conduction perpendicular the plane of the graphene in conduction perpendicular to plane (CPP) geometry. It was found that graphene was sufficient to reduce the exchange coupling between the magnetic electrodes. We also found that in a NiFe/Au/Graphene/NiFe stack, the graphene channels the spin current perpendicular to the plane and thus produces an enhanced MR effect compared to a simple stack without the graphene. A significant anisotropy was found in the observed magnetoresistance in the cross electrode geometry employed.

Index Terms—Conduction perpendicular to plane (CPP), magnetoresistive device, permalloy, spin valve.

I. INTRODUCTION

SPINTRONICS is a rapidly growing field where the spin degree of freedom is utilized in solid state systems. Spintronic devices can be much smaller, more energy efficient and more powerful for certain applications compared to conventional charge based electronics. The advantage of spin over charge is mainly due to the fact that it can easily be controlled by externally applied magnetic field and has a relatively large coherence length where unlike charge it does not dissipate with scattering or collisions over short distances. The success of spin based electronics is therefore dependent on the technology and availability of materials that possess such characteristics.

Graphene is the thinnest material known to man. It is a mono atomically thin 2-D film of Carbon atoms arranged laterally in a honeycomb benzene ring like structure. It was previously considered to be a physically unstable form until it was shown to exist in the free state [1]. 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 and room temperature carrier mobilities of $\sim 25\,000$ cm²/Vs are routinely observed. These mobilities were found to be weakly temperature dependent, which means that if impurity scattering was reduced, $\sim 200\,000$ cm²/Vs mobilities could be achieved. Transport in this system is ballistic and carriers can travel submicrometer distances (~ 300 nm) without scattering [1]–[3].

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 temperature [5]. Collectively these factors not only make graphene a novel material for electronics but also 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) [6]–[8]. However, no study has been done yet to employ graphene in devices where the conduction is perpendicular to plane (CPP). Here we look into the use of graphene as a nonmagnetic spacer layer, in multilayer spin-valve devices. Doing this we were able to successfully sandwich for the first time ever a mono-atomic thick film between metallic/magnetic thin films.

II. FABRICATING THE DEVICE

The general procedure for fabricating a device can be classified into three steps. Initially the sample substrate is patterned with a bottom electrode matrix. Then graphene is deposited onto the matrix and then a final step is carried out to pattern the top structure.

A. Electron Beam Lithography for the Bottom Matrix

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 along with reference alignment marks for relocating and aligning purposes. This particular thickness of oxide provides the best optical contrast to see graphene films [9]. This pattern is made sufficiently dense, covering a large portion of the substrate to increase the probability of obtaining some graphene flake lying on an element in the matrix. A standard liftoff technique is used to obtain the matrix after sputtering the film in the material to be used for the bottom electrode. Fig. 1(a) shows a part of this grid structure on the substrate.

B. Depositing Graphene

Graphene is obtained by repeated peeling of ~ 1 mm² flakes of natural graphite. These flakes are then transferred onto the substrate. The flakes of graphene attach themselves to the substrate by means of van der Waals forces [1]. The substrate is then inspected under an optical microscope to detect weak but visible changes in optical contrast which allows location of monolayer graphene flakes. Fig. 1(a) shows a thin flake with areas of single-, bi-, and three-layer graphene. Care is taken to remove any contamination before depositing the films. The substrate is

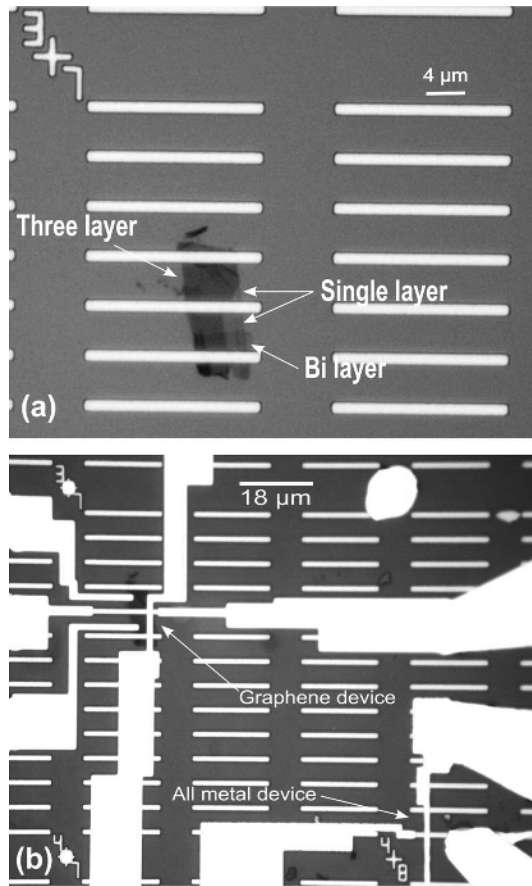


Fig. 1. (a) Optical image showing the repeated line matrix structure designed by e-beam lithography. Graphite flakes are peeled and then deposited on this structure. Optical contrast is used for identification of graphene flakes [9]. (b) Complete device structure. The top electrodes are again designed with e-beam lithography together with contacts for measurement purposes. An all metal test device was also made for comparison. The bottom grid structure is contacted with electrodes to allow a potential difference to be applied across the multilayer spin valve.

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.

C. Electron Beam Lithography for Top Layer

After identifying a graphene flake another step of e-beam lithography is carried out to design the top layer. Optical images of the film and the adjacent alignment marks make it possible to easily locate and accurately align the top electrode design for lithography. This lithography step also involves producing contacts to the upper and lower electrodes for measurement purposes. Fig. 1(b) illustrates the complete device structure after this step. A test device without graphene is included for comparison. The resultant device is then set in a chip carrier and the electrodes are contacted using gold wire (20 μm diameter) connections.

III. EXPERIMENTAL TECHNIQUES

In initial experiments the bottom layer consisted of sputtered permalloy ($\text{Ni}_{80}\text{Fe}_{20}$) on to which graphene flakes were deposited. The top layer of permalloy was then sputtered. Our measurements showed that the contact resistance to the flake

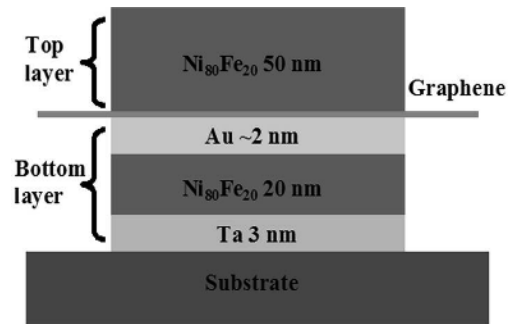


Fig. 2. Schematic cross-sectional view of the device with a Au capping layer on the bottom contacts. A 3 nm Ta layer is deposited as an adhesion layer.

was high. This was due to the fact that permalloy is highly prone to oxidation and a thin layer of oxide was formed before graphene was deposited. The nature of the preparation technique especially with regards to graphene deposition made it inevitable for the bottom layer to oxidize before graphene was deposited.

In order to avoid very high contact resistances with these devices a very thin ~ 2 nm capping layer of gold was in-situ sputtered over the bottom permalloy layer before it was removed from the deposition system. This reduced the contact resistance significantly and most of our devices were prepared in this way. Fig. 2 presents a schematic cross section of the devices used in these measurements.

IV. MEASUREMENTS AND RESULTS

Simple cross electrode resistance measurements were made for the device with graphene and without graphene as shown in Fig. 3(a) and (b), respectively. 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.

The devices (with and without graphene) were simultaneously fabricated on the same substrate. Their close proximity ($< 100 \mu\text{m}$) gives strong support for the identical duplication of the preparation conditions. Furthermore, we obtained similar results when two more experiments were repeated under the same conditions.

A thin oxide layer in the case of the device without the Au capping layer 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. 3(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 or domain wall scattering as the film switches. However, with the flake there was a stable high resistance state at zero magnetic field indicating a nonparallel magnetization state between upper and lower layers. Without the graphene flake the films appear to be sufficiently coupled to prevent magnetic reordering 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 effect. Efficient magnetic coupling in plain devices is probably related to

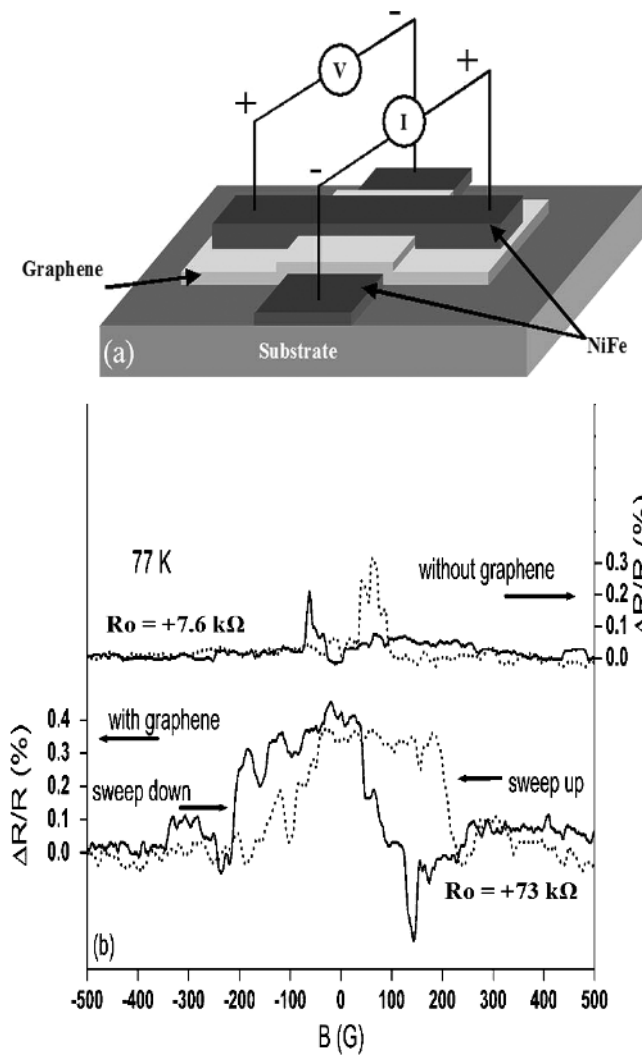


Fig. 3. (a) Schematic diagram of the device showing the graphene flake sandwiched between the layers of ferromagnetic electrodes and the configuration of the electrodes used for all measurements. (b) Magnetoresistance (MR) response of the device using only permalloy electrodes with and without the graphene flake at 77 K using simple cross electrode measurements.

pinholes 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 observed tunnel like behavior in this case, we believe this is due to the oxide layer on the surface of bottom NiFe film.

To overcome the problem of the oxide layer the bottom permalloy contacts were capped with a Au layer approximately 2 nm thick. We find qualitatively different behavior for devices with graphene and without graphene. A negative voltage was obtained from the output terminals in the case of the device without graphene. The negative sign is expected for a dominant CIP resistive component [Figs. 4(a) and 3(a)] 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. 4(a) right). The positive voltage is observed for the device with the graphene flake. This indicates that the resistive component is now more

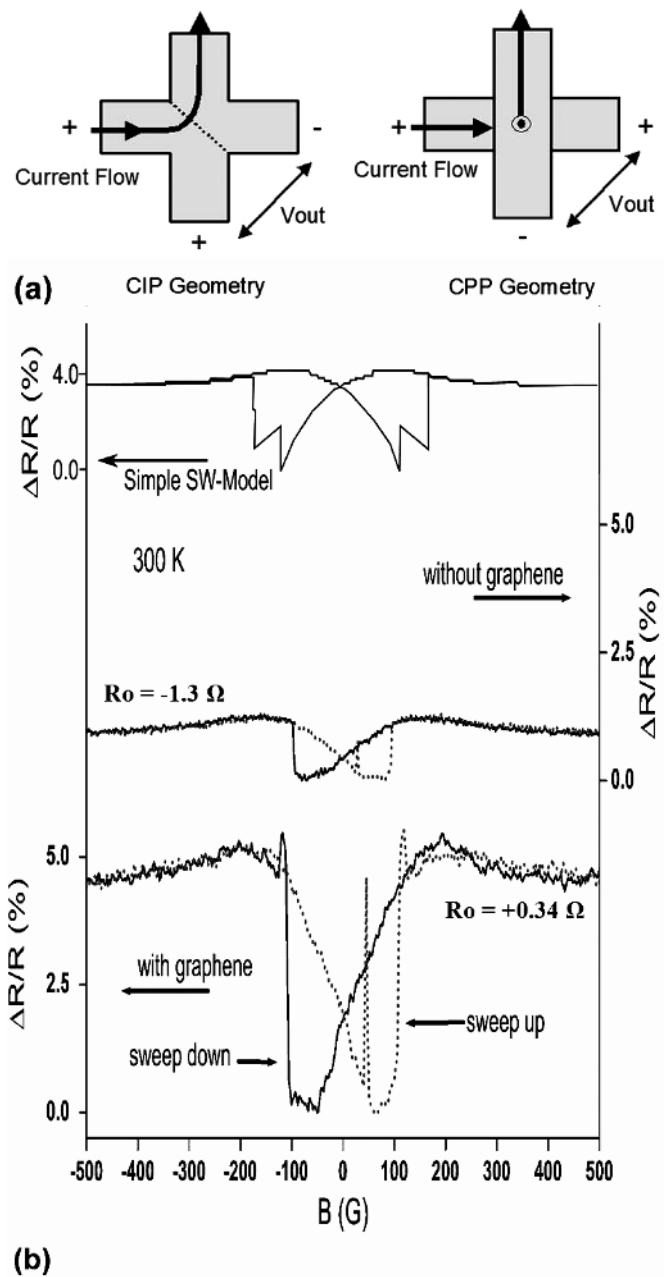


Fig. 4. (a) Schematic diagram illustrating the observed reversal of the output potential V_{out} corresponding to the CIP and CPP geometries. (b) MR response derived from the absolute values of the resistance measured for the device with the Au capping layer with a graphene layer and without graphene. 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.

dominated by the current flowing perpendicular to the device plane (CPP geometry). Fig. 4(a) illustrates this by showing the extreme cases for the current path with and without the graphene flake present. The I - V curves in this case were found to be linear with and without graphene. Consequently, tunneling is thought to play no significant role.

Fig. 4(b) shows the percentage MR response derived from the resistance measurements. Absolute values have been taken ignoring the sign difference between the devices. An MR response was seen in both cases. In the device with graphene, the MR response was larger by a factor of four. In a standard spin valve

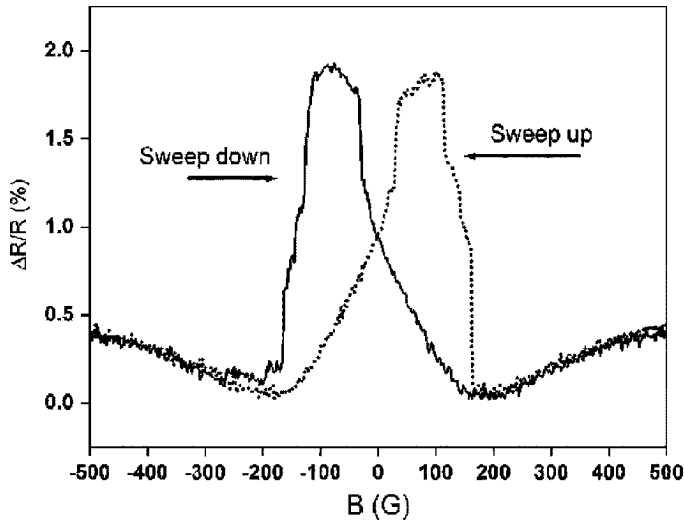


Fig. 5. MR response taken on device without Graphene and with the applied field in a direction orthogonal to that for the curves in Fig. 4. Comparing this with Fig. 4 shows the degree of anisotropy in the measured MR.

response it would be expected that at higher field there would be parallel magnetization between the top and bottom layer and hence lower resistance. There is significant anisotropic MR as can be seen from the curves shown in Fig. 5 taken with the field orthogonal to that of Fig. 4. It is clear from these figures that there is a significant negative MR component, which depends on the angle of the applied field and is similar to the results obtained by Suezawa and Gondo for Ni/Co cross junctions [10].

A simple model has been developed with the crossed conductors modeled as two Stoner-Wohlfarth ellipsoids with widths approximating to the conductor widths. The MR was computed by assuming a positive AMR component varying as $\cos^2(\varphi)$, where φ is the angle between the magnetization and the current flow direction in each conductor. To this was added a negative GMR component varying as the cosine of the angle between the magnetization directions in the ellipsoids. An AMR of 2% and a GMR of 4% were assumed. The free energy was minimized at each applied field value and the resultant resistance was computed. As can be seen from the top curve in Fig. 4(b) the general shape of the experimental MR dependence is reproduced, but to obtain a better fit to the switching field and predict the detailed shape more accurately a full micromagnetic model would be required.

V. CONCLUSION

We believe that the current is being channeled through graphene, which is evident from the change in sign of the measured voltage on the cross. This channeling is probably taking place at defects within the area of the graphene flake sandwiched between the top and bottom layer.

We conclude that the increase in the MR is due to the CPP conduction becoming more dominant with the graphene in place unlike the device without the graphene flake where the CIP component dominates. In the case of devices without Au capping layer we observe that a single layer flake of graphene is sufficient to significantly weaken the exchange coupling between two ferromagnetic layers.

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