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### Communication

## Influence of DC-biasing on the performance of graphene spin valve

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#### ABSTRACT

Generating and controlling the spin valve signal are key factors in 'spintronics', which aims to utilize the spin degree of electrons. For this purpose, spintronic devices are constructed that can detect the spin signal. Here we investigate the effect of direct current (DC) on the magnetoresistance (MR) of graphene spin valve. The DC input not only decreases the magnitude of MR but also distorts the spin valve signal at higher DC inputs. Also, low temperature measurements revealed higher MR for the device, while the magnitude is noticed to decrease at higher temperatures. Furthermore, the spin polarization associated with NiFe electrodes is continuously increased at low DC bias and low temperatures. We also demonstrate the ohmic behavior of graphene spin valve by showing linear current-voltage (I-V) characteristics of the junction. Our findings may contribute significantly in modulating and controlling the spin transport properties of vertical spin valve structures.

#### 1. Introduction

Spintronics, utilizing the spin degree of electrons is thoroughly studied since from the last two decades [1-15]. Owing to its importance, it is highly desired to integrate spintronic devices with tunable characteristics. One such type of device is vertical spin valve, which is integrated by inserting a non-magnetic spacer between the two ferromagnetic (FM) electrodes. This type of configuration forms current-perpendicular-to-plane (CPP) geometry in which current flows perpendicular to the plane of device. The devices hold great potential for various kinds of applications including information storage and spin based logic [3,16–22]. Therefore, controlling the spin valve signal (that is defined as the relative magnetoresistance (MR), MR= (RAP - $R_P$ / $R_P \times 100\%$ ) is essential feature in these magnetic junctions. Many attempts have been made to modulate the MR of such spin valve devices [14,22-25]. For instance, graphene was directly grown on the bottom FM electrode to overcome the surface oxidation and contaminations at FM/graphene interface; thereby to improve the spin properties. Relatively higher MR ratios were achieved due to improved fabrication processes [9,10,26]. A similar effort was made by M. Piquemal-Banci et al., wherein hBN was directly grown on the FM film [23]. Their results were quite modified as compared to previously reported results pertinent to

In this paper, the magnetoresistance (MR) is controlled through DCbiasing. A monotonic decrease is observed in the magnitude of MR and the signal show fluctuations as the value of DC is increased beyond some critical value. Temperature dependent measurements are also performed, which revealed a monotonic increase with decreasing temperature. The spin polarization of FM leads decreases with increasing DC-biasing and temperature, respectively. Our approach opens a new way for controlling the spin related properties of vertical magnetic architectures.

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MR and spin polarization of FM leads [27,28]. Because of the exposure of bottom FM layer to ambient environment, the surface oxidation of the electrode is inevitable. A thin Au layer was deposited on the bottom FM film to prevent oxidation [14,29]. As expected, the magnitude of MR was considerably raised owing to the reduced oxidation at the surface of bottom FM electrode. The effect of different FM metals and the quality of the intervening layer on the spin transport properties was also investigated [4,30]. Moreover, AC-biasing has been thoroughly studied for various vertical magnetic junctions incorporating different non-magnetic interlayers [4,31]. However, the effect of DC-signal on the magneto-transport properties is hardly reported so far. Therefore, it is very crucial to investigate the influence of DC biasing on the spintronic properties of spin valve architectures.

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**Fig. 1.** (a) The schematic of graphene spin valve, which shows the general structure of the device and the mechanism for transport measurement. (b) Raman spectrum of graphene revealing good quality for the layer. The inset illustrates the optical image of the fabricated device. (c) Current-voltage characteristics for graphene spin valve showing metallic behavior.

#### 2. Experimental details

Graphene flakes were well grown on copper foils using the wellknown chemical vapor deposition (CVD) technique as described in our previous papers [32,33]. After growing graphene on the Cu foil, it was transferred to NiFe electrode to form the device. The graphene growth and transfer processes are explained in ref. [12]. Spin valve was fabricated by depositing NiFe electrode on Si substrate using e-beam lithography before transferring graphene. After transferring graphene film, NiFe was again patterned on the top of graphene to form current perpendicular to plane (CPP) geometry by using the as mentioned e-beam lithography and the lift-off procedure. To avoid the bypass current, the removal of extra graphene layer was essential. It was attained through O<sub>2</sub> and XF<sub>6</sub> plasma etching method. We performed the four probe electronic transport measurements by utilizing the famous ac lock-in technique by operating the system at a fixed frequency of 11.7 Hz, which delivered a current of 50 µA. In order to carry out the low temperature measurements, liquid helium and Lake Shore 331 temperature controller were utilized for cooling the samples.

#### 3. Results and discussion

A vertical spin valve device is formed when any two FM leads are contacted through a non-magnetic interlayer. If the resistance of device is measured by sweeping magnetic field that is obliquely applied to the device plane, two distinct states of resistance arise. First, the low resistance situation appears, wherein the magnetization vectors of two FM leads are aligned parallel to each other. This state is favorable for spin polarized electrons to pass easily from the junction; the spin current is maximum in such condition. Second, high resistance of the junction emerges when the spins of FM leads are oriented anti-parallel. Such combination of the resistance states gives rise to spin valve effect. Fig. 1a illustrates the schematic structure of graphene spin valve in which two NiFe electrodes are decoupled by graphene layer. The mechanism of transport measurement is also depicted in schematic i.e. four probe

measurement is utilized. The circle enclosing plus-minus indicates DCbiasing, which is the topic of this investigation. Raman spectroscopy is usually employed for characterizing two-dimensional (2D) materials [34–38]. Here, we utilized it to determine the thickness of our graphene sample. Fig. 1b exhibits the Raman spectrum of the graphene layer representing characteristic peaks i.e. G and 2D at  $\sim 1586 \text{ cm}^{-1}$  and  $\sim$ 2691 cm<sup>-1</sup>, respectively. The spectrum reveals small intensity for D peak indicating good quality of graphene film. Furthermore, the intensities ratio  $(I_{2D}/I_G)$  is estimated to be ~5.7, which confirms single-atom thickness for graphene. In order to create shape anisotropy in the ferromagnetic electrodes, the width of FM1 was taken as  $0.78\,\mu m$ while that of FM2 it is taken to be 3.3  $\mu m$  . Therefore, each ferromagnetic electrode should reveal different switching fields of magnetization. The junction area is found to be  $2.574 \,\mu m^2$ . The interface resistance for the junction is calculated to be  $\sim$ 20.33  $\Omega$  at 50 K and  $\sim$ 38.26  $\Omega$  at 300 K. The real optical micrograph of the device is also displayed in the inset of Fig. 1b, which shows that FM electrodes are decoupled with the interlayer. Also, the direction of applied magnetic field is pointed in the inset. The I-V measurements are performed at two different temperatures for vertical graphene magnetic junction to understand the behavior of graphene based magnetic junction, which shows linear I-V characteristic curves indicating metallic behavior of graphene for vertical electrical transport (Fig. 1c). This exhibits the formation of ohmic junction and elucidate the fact that charge transport does not occur through tunneling mechanism.

Spin valve effect is defined in terms of relative MR ratio, which can be calculated as MR=  $(R_{AP} - R_P)/R_P \times 100\%$ . Here,  $R_{AP}$  corresponds to B-field dependent resistance, while  $R_P$  refers to resistance when the two FM leads are magnetized in parallel configuration. Fig. 2a elucidate the results of MR ratio for the graphene spin valve at different DC inputs and 50 K temperature. The magnitude of MR is calculated to be approximately 0.24% at 50 K, which is smaller than our previous results at room temperature [26]. It is due to the reason that we performed direct growth of graphene on Ni electrode, which prevented surface oxidation at Ni electrode and thus improved the interface quality. It can be seen that at

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