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Research Article

Magnetoresistance Effect in NiFe/BP/NiFe Vertical Spin Valve Devices

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Two-dimensional (2D) layered materials such as graphene and transition metal dichalcogenides are emerging candidates for spintronic applications. Here, we report magnetoresistance (MR) properties of a black phosphorus (BP) spin valve devices consisting of thin BP flakes contacted by NiFe ferromagnetic (FM) electrodes. The spin valve effect has been observed from room temperature to 4 K, with MR magnitudes of 0.57% at 4 K and 0.23% at 300 K. In addition, the spin valve resistance is found to decrease monotonically as temperature is decreased, indicating that the BP thin film works as a conductive interlayer between the NiFe electrodes.

1. Introduction

Two-dimensional (2D) nanomaterials such as single-layer graphene and transition metal dichalcogenides (TMDs) have attracted great attention as building blocks for future (opto)electronic technologies due to their specific layered structures and novel physical properties [1–3]. Recently, the 2D nanomaterials have also been demonstrated to have potential for application in the field of spintronics [4–11]. The 2D materials have been largely researched as nonmagnetic interlayer of spin valve, which is similar to traditional magnetic tunneling junctions consisting of two ferromagnetic (FM) layers separated by a nonmagnetic insulating spacer, usually Al_2O_3 and MgO, and the resistance depends on the magnetization orientation of two ferromagnetic electrodes [12, 13]. The first experimental work for realization of spin-transport phenomenon is reported by Tombros et al. in graphene-based planar spin valve structure [4]. Later on, magnetoresistance (MR) was measured at room temperature in graphene vertical spin valve [7, 8]. Subsequent works reported spin-dependent transport in h-BN and transition metal dichalcogenides (TMDs), such as MoS_2 and WS_2 [9–11]. These studies suggest that the 2D nanomaterials may be promising for spintronic applications. Recently, a few-layer black phosphorus (BP), a newly identified 2D nanomaterial, has been demonstrated to be an appealing candidate material owing to its exotic physical properties such as thickness-dependent tunable band gap and high carrier mobility [14–18]. Interestingly, theoretical study predicted a relatively large MR ratio in the BP-based spin valve structure [19–21]. However, so far, there have been no reports of MR effect in BP-based spin valve.

In this work, we report on the fabrication and spin valve effect in the BP-based device. 2D BP is sandwiched by two permalloy electrodes (Py and $Ni_{81}Fe_{19}$) and the BP layer serves as nonmagnetic spacer layer as shown in Figure 1(a). The devices show spin valve effect from room temperature to low temperature with a MR of 0.57% at 4 K. The temperature dependence of the device resistance reveals that the BP layer works as a metallic layer between two FM electrodes.

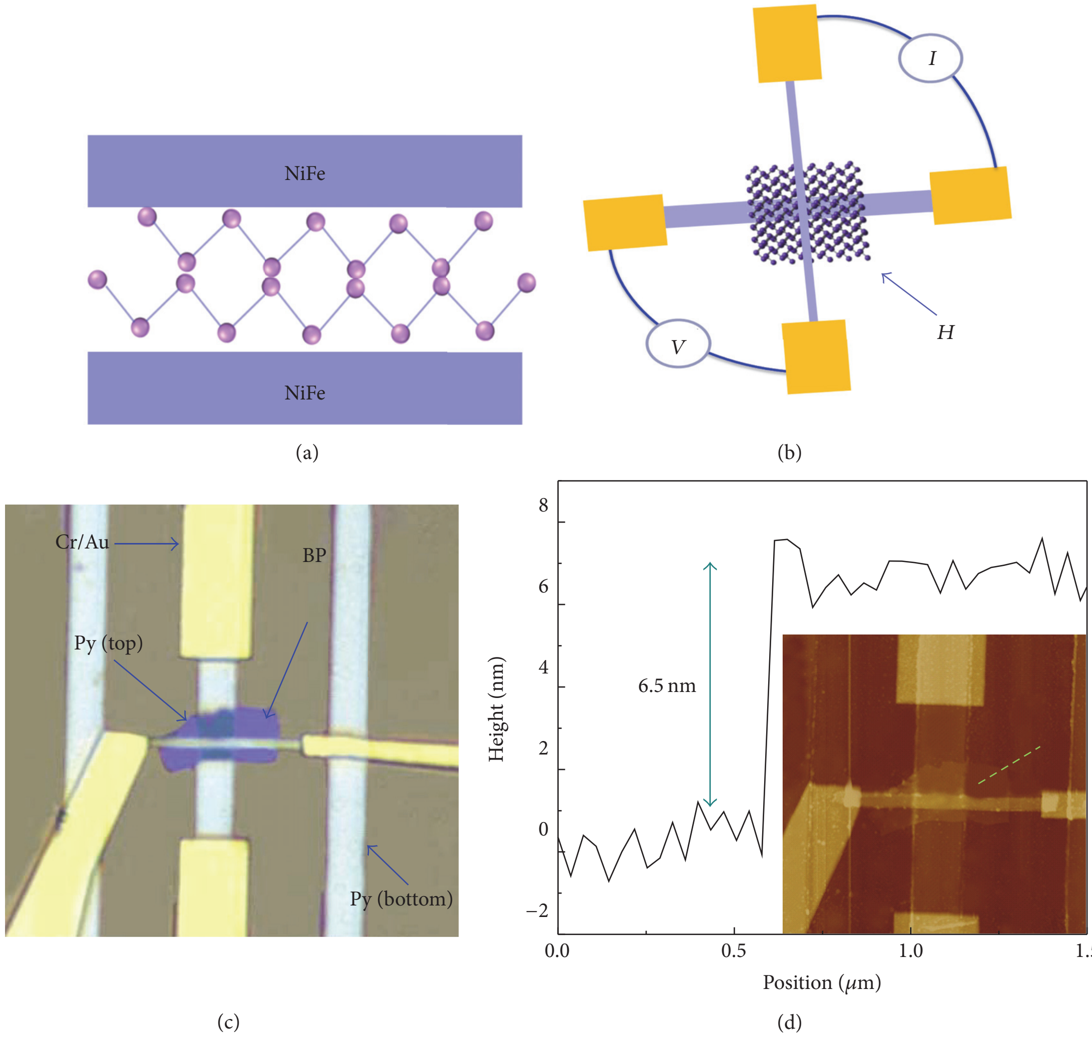

FIGURE 1: ((a) and (b)) Structure and measurement structure of BP-based vertical spin valve device, consisting of bottom NiFe electrode, 2D-BP spacer, and top NiFe electrode. (c) Optical macrograph of NiFe/BP/NiFe spin valve device. (d) AFM graph of device.

2. Methods

2.1. Material and Device Fabrication. The BP crystals were synthesized from red phosphorus under high temperature of 1000°C and high pressure of 2 GPa. Thin BP flake was obtained by mechanically exfoliating BP crystal using adhesive tape (scotch tape), and then the flake was transferred onto the prepatterned Py (bottom) electrodes on SiO_2/Si substrate. The bottom electrodes were fabricated by e-beam lithography (EBL) and a lift-off procedure after e-beam evaporating Py with thickness of ~30 nm. In the subsequent process, top Py electrodes with thickness of ~50 nm were fabricated by another run of EBL, metal deposition and lift-off process. Finally, the bottom and top FM electrodes were connected with large electrodes by EBL and Cr (5 nm)/Au (60 nm) deposition.

2.2. Device Characterization and Measurement Setup. The devices were measured with a four-terminal setup, where the bias currents flow perpendicular to the device plane of the spin valve. The magnetic field was applied in-plane at 45° to the direction of the Py ferromagnetic electrodes as shown in Figure 1(b). The BP flake was initially identified by optical microscopy in Figure 1(c) and then further confirmed by atomic force microscopy (AFM). Figure 1(d) shows an atomic force microscope (AFM) image of one device, revealing that the thickness of BP for the device is ~6.5 nm. The transport measurements for the BP-based spin valve devices were performed using Physical Properties Measurements System (PPMS) made by Quantum Design.

3. Results and Discussion

3.1. Current-Voltage Characteristics and Spin Valve Effect. Figure 2(a) displays the current-voltage (I-V) curves of one typical device for various temperatures. The linear I-V curves indicate the Ohmic contact characteristics of the BP flake and FM electrodes. At room temperature, the resistance-area

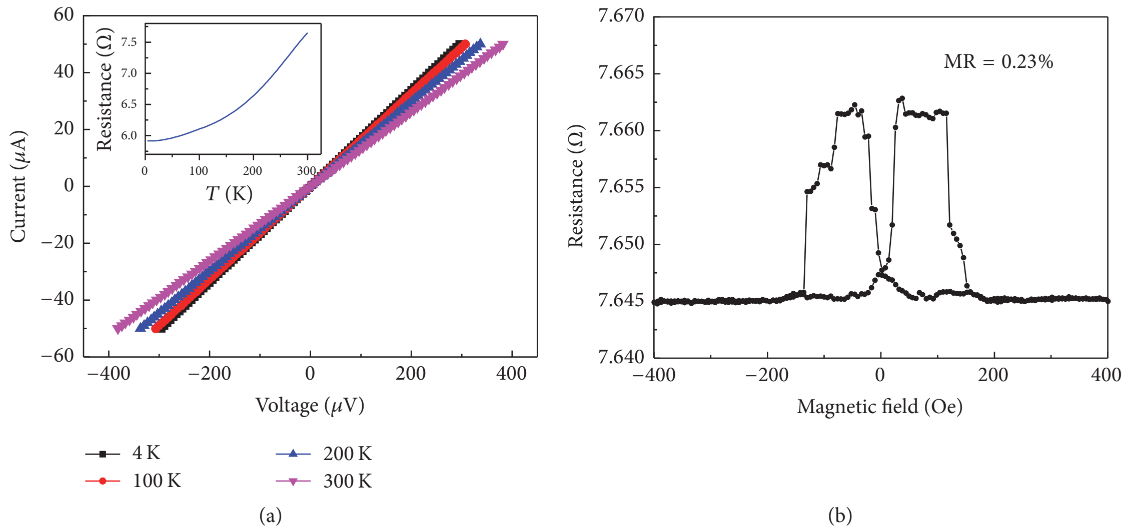

FIGURE 2: Characterizations of NiFe/BP/NiFe spin valve device. (a) The current-voltage curves of device at different temperature from 4 K to 300 K and resistance curve as a function of temperature at zero magnetic field in inset figure (a). (b) Resistance curves of device when external magnetic field is applied at 300 K and the value of MR is 0.23% for the structure of Py/BP/Py spin valve.

(RA) product of the device is on the order of $\sim 10^{-11}\ \Omega\cdot\mathrm{m}^2$, which is smaller than that in the monolayer MoS_2 device ($\sim 10^{-10}\ \Omega\cdot\mathrm{m}^2$ [10]). This difference may be related to the different band gaps (~0.5 eV in our device while ~1.87 eV in [10]). Note that the resistance decreases with reducing temperature, indicating that the BP behaves as a metal in the spin valve structure as shown in inset of Figure 2(a). The results suggest that the thin BP flake behaves as a conducting thin film rather than a tunnel barrier between the two FM electrodes, which is consistent with the previous works in MoS_2 and WS_2 based spin valve [10, 11].

Spin valve effect is characterized by measuring the resistance as a function of magnetic field. By sweeping the magnetic field, the resistance can be tuned into the high-resistance (R_{AP}) state and low-resistance (R_{P}) state since it depends on the orientation of the magnetization of the FM electrodes. The magnetoresistance is defined as $\mathrm{MR} = 100 \times (R_{\mathrm{AP}} - R_{\mathrm{P}})/R_{\mathrm{P}}$, where R_{AP} and R_{P} are the resistances when the magnetization vectors of two Py electrodes are antiparallel and parallel to each other, respectively. Note that the widths of the top and bottom electrodes were designed to be 500 nm and 2 μm as shown in Figure 1(c), respectively. This yields a large difference in coercivity between two FM electrodes. Thus the bottom electrode is easier to magnetize than the top one under the application of magnetic field owing to weaker shape anisotropy. The resistance as a function of magnetic field for a representative Py/BP/Py spin valve at room temperature is shown in Figure 2(b). As the magnetic field scan from −400 Oe to 400 Oe, the top and bottom electrodes switch in sequence, resulting in the observation of a resistance plateau. The MR value of the device is determined to be 0.23% at RT.

3.2. Temperature Dependence of the Spin Valve Effect. Figure 3(a) shows a series of MR curves for a representative Py/BP/Py spin valve at various temperatures ranging from 4.2 K to 300 K. The maximum MR value is 0.57% at 4 K. A simple relation between the MR and the polarization of the FM electrodes for a junction can be approximated as $\mathrm{MR} = 2P_1P_2/(1 - P_1P_2)$, where P_1 and P_2 are the electron spin polarization of the two FM metals, respectively [10]. Assuming that two FM electrodes have the same composition, then the polarization ($P_1 \approx P_2 = P$) of the two Py electrode is estimated to be ~5%, which is comparable to the other values reported previously [10], but smaller than that ($P \sim 0.3$) in the Py/Al_2O_3 interface. A possible reason for this reduced polarization is the exposure to air of the Py surface prior to the application of the BP layer. Some contaminants are inevitably adsorbed on the surface and air exposure of Py may produce antiferromagnetic NiO, significantly decreasing the spin polarization [22]. Future well-controlled fabrication process in situ without air exposure of the interfaces may improve the interface quality, maximizing the MR effect.

The magnitude of the MR monotonically decreases as the temperature is increased, as shown in Figure 3(b). The decrease in MR amplitude at higher temperature may be attributed to the inelastic scattering with phonons, magnetic impurity scattering, surface states, and thermal smearing of electron energy distribution in the FM metals [23]. The data are found to follow Bloch's law, where the spin polarization is described by $P(T) = P(0)(1 - \alpha T^{3/2})$. By fitting the data with MR relation by considering the temperature dependence of spin polarization, the material-dependent constant α can be estimated to be $5.9 \times 10^{-5}\ \mathrm{K}^{-3/2}$. This value is comparable to that of $3\sim5 \times 10^{-5}\ \mathrm{K}^{-3/2}$ reported in the literature [24]. Temperature-dependent relation of parallel and antiparallel resistance are showed in Figure 3(c), which further indicated BP not as insulting barrier layer in the MTJ, but as metallic layer in vertical spin valve.

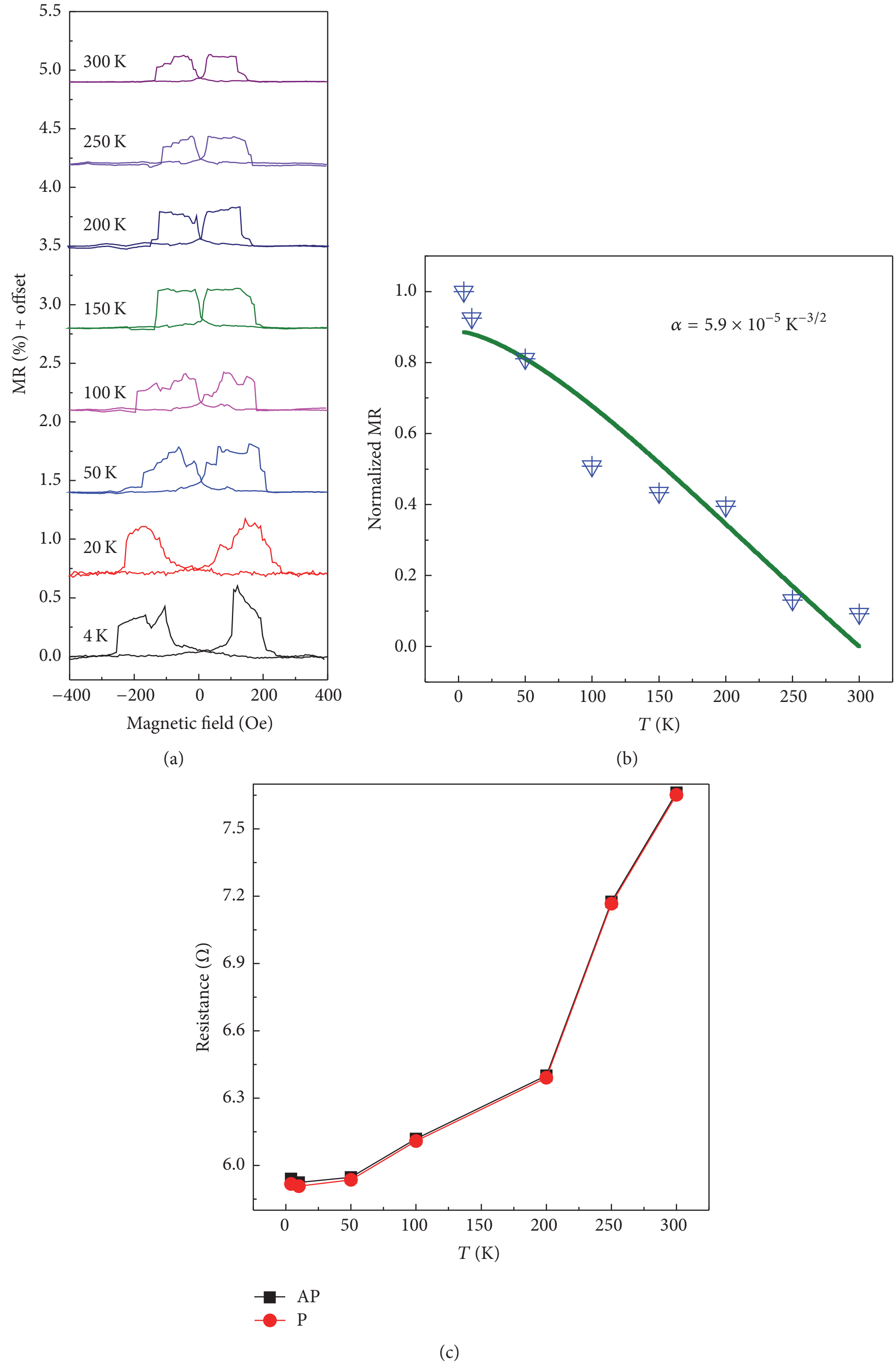

Figure 3: Temperature dependence of spin valve effect in NiFe/BP/NiFe device. (b) Normalized MR ratio as a function of temperature and the solid line is the fitting to Bloch's law. (c) Resistance of device, corresponding to low resistance (R_P) and high resistance (R_{AP}).

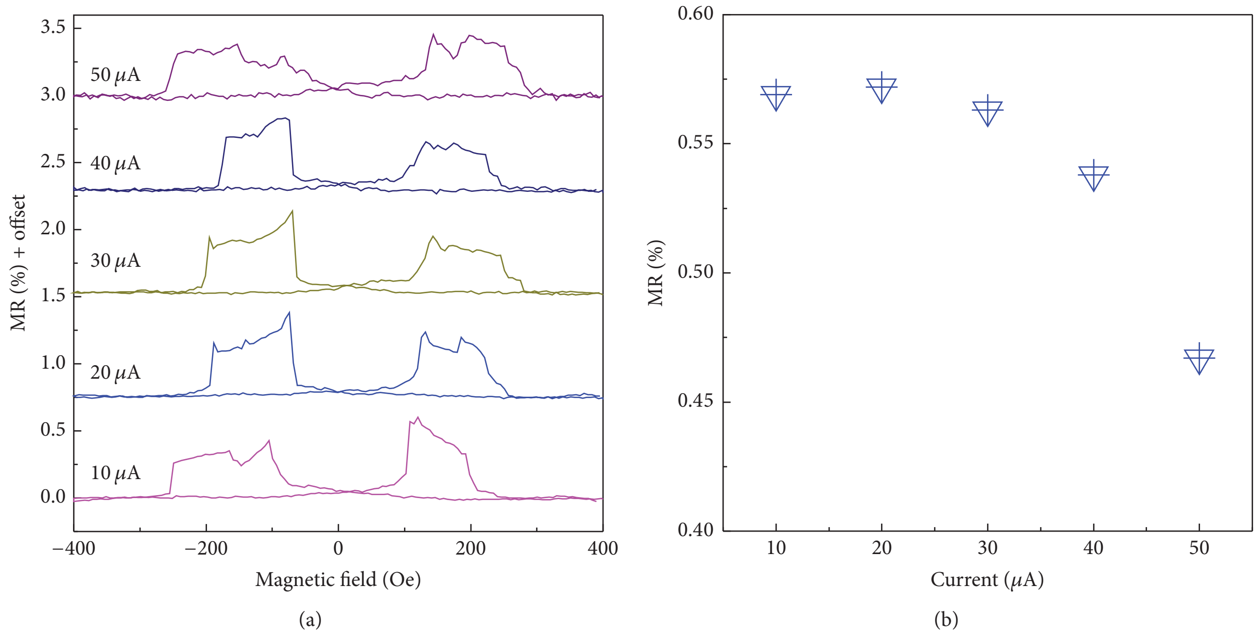

FIGURE 4: (a) Magnetoresistance curve at various bias currents. (b) MR ratio as a function of bias current.

3.3. Bias Current Dependence of the Spin Valve Effect. Finally, we investigated the bias current dependence of the spin valve effect. Figure 4(a) shows the resistance as a function of magnetic field at various bias currents from 10 μA to 50 μA at 4 K. The amplitude of MR value is found to be decreased as the bias current increases as shown in Figure 4(b). We attribute the decrease in MR value at larger bias current to the spin excitations localized at the interfaces between the FM electrodes and the BP interlayer [25] as well as the localized trap states in the BP interlayer [26] at the BP interlayer played a metallic property role in vertical spin valve. The results provide a possible approach to use the emerging BP nanomaterials for future spintronics applications such as magnetic memory and logic devices.

Competing Interests

The authors declare that they have no competing interests.

Acknowledgments

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