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Permalloy and $Co_{50}Pd_{50}$ as ferromagnetic contacts for magnetoresistance measurements in carbon nanotube-based transport structures

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In this paper, magnetoresistance (MR) measurements on carbon nanotube (CNT) 2-terminal spin-valve devices are presented. Results from samples with both permalloy (Py) and CoPd contacts show repeatable MR switching. In performing gate-dependent MR measurements on the Py-contacted CNTs, two distinct transport regimes are identified, and their transport behavior is discussed with respect to optimizing MR. Results from the first CoPd-contacted CNTs indicate a stable magnetic response with a higher magnitude than that of a Py-contacted nanotube in the same transport regime. © *2012 American Institute of Physics*. [doi:10.1063/1.3673841]

I. INTRODUCTION

In addition to exhibiting ballistic transport, carbon nanotubes (CNTs) are thought to have small spin-orbit interaction and relatively few spin nuclei (^{13}C), suggesting a long spin relaxation length. This makes CNT-based devices interesting for studying spin-dependent transport and potential applications in the field of spintronics, especially as electric-field control of the magnetic behavior of samples has been observed.[1,2]

In order to create a reliable device, however, the spin injector and detector, in this case ferromagnetic contacts, must be optimized to have both high in-plane magnetic polarization, and form a stable electronic interface with CNTs. Previous work in this field has focused on obtaining and understanding magnetoresistance (MR) in the single-electron tunneling (SET) transport regime.[1,3] It is well established that low-temperature transport in the SET regime in CNTs occurs via tunneling through a potential barrier that develops between the CNT and a metallic contact.[4] Therefore, MR in CNT-based devices is considered to be tunneling magnetoresistance, and the nanotube-contact interfacial properties largely determine the ability to achieve and control MR. In this work, we investigate how the MR changes when devices are driven beyond the SET regime. We compare two materials, the well-studied weak ferromagnet permalloy (Py - $Ni_{81}Fe_{19}$), and an alloy of cobalt and palladium, which has a more complex magnetic behavior, but is expected to have a higher polarization,[5] and form more transparent contacts to CNTs. The aim of the work is to obtain a high, reliable MR outside the SET regime, all of which are criteria for eventual applications.

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II. EXPERIMENTAL DETAILS

In this work, CNTs were grown via chemical vapor deposition onto lithographically prepatterned Si/SiO_2 substrates. The silicon was highly doped for use as a back gate. Methane was used as a carbon precursor gas, and the catalyst was a standard Fe_2O_3-based solution. A growth temperature of approximately 920 °C was used, which we expect to grow mainly single and double-walled CNTs.[6] The CNTs were subsequently located via atomic force microscopy (AFM) and contacted by electron-beam lithography. E-beam lithography and metal evaporation were performed in two steps. First, ferromagnetic contacts were deposited and capped with 5 nm of gold. 40 nm thick Py contacts were evaporated as an alloy, while Co and Pd were coevaporated via molecular beam epitaxy to produce 25 nm thick $Co_{50}Pd_{50}$ contacts. In the next step, larger gold contacts and bond pads were deposited via e-beam evaporation.

The architecture of typical samples is shown in Fig. 1. For 2-terminal measurements, a drop in resistance between two consecutive contacts with respect to external magnetic field sweep was measured. It can be seen that for Py-contacted samples, longer, rectangular contacts were used, with dimensions of approximately 3 $\mu m \times 150(400)$ nm for the narrower (wider) contacts. As we have previously reported, CoPd alloys exhibit multiple magnetic domains in such geometries.[7] We therefore simplified the micromagnetics of the system by creating smaller, oval contacts with the long axis being no more than 2 μm. Both samples analyzed in this paper appeared metallic at room temperature, with $R = 150$ kΩ for sample (a) and 50 kΩ for sample (b).

All transport measurements presented here were taken at 4 K in a He^{4+} flow cryostat. The magnetic field was always swept in-plane to the sample, along the contacts' long axes. The sample shown in Fig. 1(a) was exposed to only one thermal cycle, whereas that shown in Fig. 1(b) was cooled down three times, explaining the difference in roughness of the

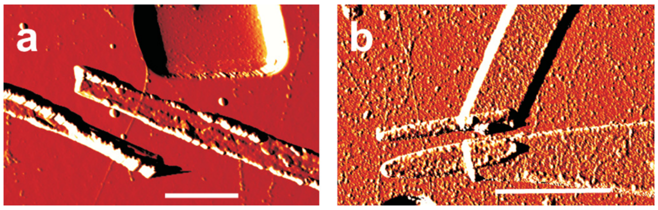

FIG. 1. (Color online) AFM images of sample architecture. Shown here are representative 2 - terminal (a) Py-CNT-Py and (b) CoPd-CNT-CoPd devices. (Scale bars, 1 μm).

samples. For all samples discussed, the strength of the MR signal is defined as:

$$MR = \frac{\Delta R}{R_{\text{parallel}}} \cdot 100\%. \tag{1}$$

III. RESULTS AND DISCUSSION

Data measured on a 2-terminal Py-contacted CNT is presented in Fig. 2. Figure 2(a), shows gate sweep measurements of the sample, where the bias voltage was constant at +/−2 mV and −5 mV. Coulomb oscillations appear in the entire sweep range, indicating the formation of a potential barrier, as expected for Py-contacted CNTs.[3] The oscillations reproduce nicely over several bias voltages, which ultimately shows the stability of the device. While room temperature measurements showed the CNT to be chirally metallic, the gate-dependence indicates the opening of a stress-induced band-gap.[8]

Two distinct transport regimes may be defined by these measurements. They appear at all bias voltages, but are most easily visualized in the sweep of $V_{bias} = -5$ mV, which will therefore be discussed here. First is the few-electron tunneling regime, which is present when a gate voltage between 0 and −1.2 V is applied, labeled as Regime I in Fig. 2(a). When $V_{bias} = -5$ mV is used, the current oscillates around zero, but is never completely suppressed, meaning the device never reaches Coulomb blockade. At this point, the sample is clearly operating in the few or multiple-electron tunneling regime.

Regime II appears at V_{gate} below −1.2 V. Here, the Coulomb oscillations are less pronounced, although still present, indicating that a tunnel barrier is still present and contributing to the transport properties of the device. However, the current remains high, rather than fluctuating back to a value around zero, as seen in Regime I. This is clearly caused by the device beginning to behave as an opening transistor, rather than simply allowing a few electrons to tunnel at a time. It is therefore necessary to consider the possibility of an additional ohmic contribution to the resistance, which can strongly influence magnetotransport, as discussed below.

Magnetoresistance measurements were then performed on the sample using a series of bias voltages representing both transport regimes I and II. First, measurements were taken at $V_{bias} = -5$ mV and $V_{gate} = 0$ and −0.4 V, points clearly in the few-electron tunneling regime. For these measurements, no MR was observed, although it is possible that MR was present, but less than 1%, and therefore lost in the signal-to-noise ratio of the measurement. Figure 2(b) shows MR measurements performed using $V_{bias} = -5$ mV and $V_{gate} = -1.37$ and −2 V respectively. Both of the latter points are in Regime II. Clear peaks in the resistance values were measured between −5 and −25 mT and 20−45 mT (a slight shift toward positive fields was observed, likely due to exchange bias), with MR = 3% for $V_{gate} = -1.37$ V and 4.5% for $V_{gate} = -2$ V.

This data provides an important addition to the current understanding of how gate-dependent MR works. It has previously been shown that for devices in the SET regime, the maximum MR signal is observed at gate voltages corresponding to a minimum in the current.[1] Our measurements show that this effect is not linear when driving the device out of the SET regime. While for small currents in Regime I, any MR present was below the detection limit, a larger signal appeared in Regime II, as the gate began to drive the device into the p-conduction region, even though the absolute current through the device was much higher. However, within Regime II, larger MR was obtained at $V_{gate} = -2$ V than at $V_{gate} = -1.37$ V, even though the absolute current was lower, in agreement with trends within the SET regime.[1] Similar results were achieved for multiple Py-contacted CNTs.

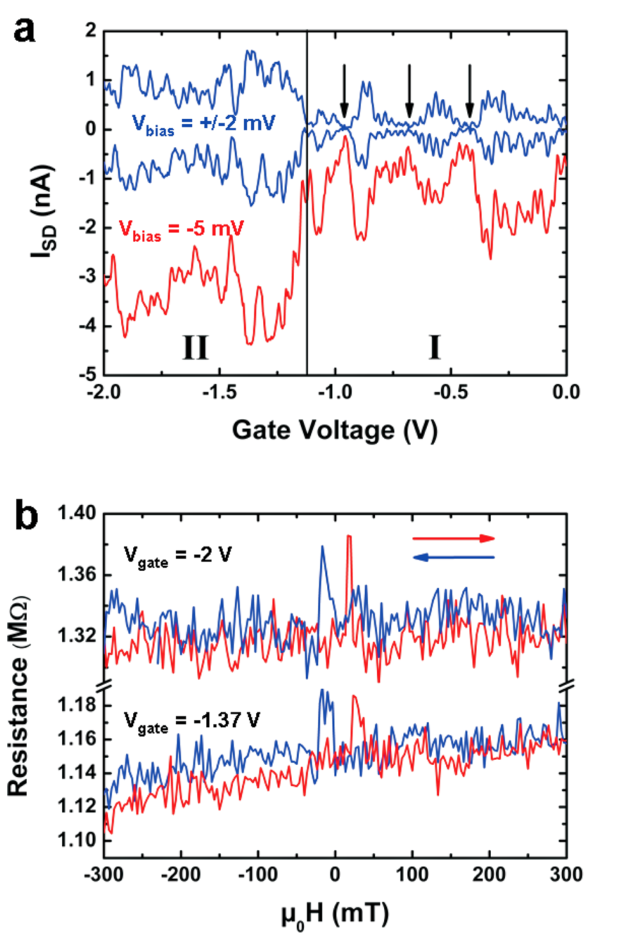

FIG. 2. (Color online) Magnetotransport in a 2-terminal Py-CNT-Py device. (a) Gate sweeps at $V_{bias} = +/-$ 2 and −5 mV. Arrows indicate regions of partially suppressed current. The line is a guide to the eye distinguishing between two transport regimes. (b) Magnetic field sweeps with $V_{bias} = -5$ mV and $V_{gate} = -1.37$ (lower curves) and −2 V (upper curves). TMR = 3% and 4.5%, respectively. Arrows indicate sweep direction.

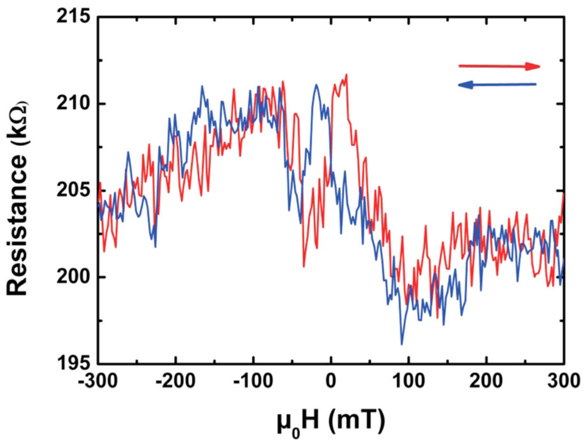

FIG. 3. (Color online) MR measurement on a CoPd-contacted sample. For this measurement, $V_{bias} = 3\,\mathrm{mV}$ and $V_{gate} = 0\,\mathrm{V}$. Arrows indicate sweep direction.

Driving a device into a regime where it acts as an opening transistor is therefore one way to increase MR in a CNT-based device. This may also be accomplished by exchanging the weak ferromagnet Py with a more suitable material. Ferromagnetic alloys based on palladium MPd (where M = Ni, Fe, Co) have been shown to exhibit a high polarization,[5] while Pd is known to create stable, transparent interfaces to CNTs.[9] FePd is expected to have the highest polarization, but has been shown to exhibit a complicated phase diagram and magnetic structure.[3] CoPd has a magnetic moment comparable to that of FePd, and has a simple fcc structure.[5] Furthermore, it has been shown that optimization of contact geometry should result in single-domain CoPd contacts.[7] For this reason, we expect CoPd-contacted CNTs to have a high, stable MR with respect to that of Py.

Figure 3 shows a magnetoresistance measurement performed on a 2-terminal CoPd- contacted CNT. After cooling down, the average sample resistance for parallel contact polarization is $R_{\parallel} = 202\,k\Omega$. This resistance, which maintains a similar order of magnitude for several CoPd-contacted devices, is significantly smaller than that of a typical Py-contacted CNT. This is likely due to the fact that Pd creates a more transparent contact to CNTs, resulting in lower potential barriers at low temperatures.

In the measurement shown in Fig. 3, applying $V_{bias} = 3\,\mathrm{mV}$ and $V_{gate} = 0\,\mathrm{V}$ resulted in MR = 5%, which already exceeds the optimized value we observed for Py-contacted CNTs. A background appears which remains constant for this device, and while not fully understood, is thought to be the result of electron-magnon interactions.[3] Gate sweeps on multiple samples did not result in any observed gate-dependence. We may therefore consider that Pd-based contacts to CNTs indeed form only low potential barriers and that the device in Fig. 3 can only be in Regime II, where we have previously achieved the maximum MR using permalloy contacts. Although the exact magnitude of MR we observe is, of course, sample specific, our results show for the first time that shaped CoPd contacts to CNTs result in reproducible, symmetric switching with a high MR appearing in a transport regime that has potential for eventual application.

IV. CONCLUSION

In summary, reliable 2-terminal quasi spin-valve devices have been fabricated by contacting CNTs with permalloy and CoPd. Gate-dependent measurements on a Py-contacted CNT show that driving the device away from the few-electron tunneling regime increases the MR. Results from CoPd-contacted CNTs give an even higher MR signal, even without using the gate or bias voltage to optimize the device performance. This report of complete, symmetric MR in CoPd-contacted nanotubes is a clear indication of the ability of the system to display stable, high-intensity magnetoresistance.

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