Large magnetoresistance in single-walled carbon nanotubes contacted different ferromagnetic metal electrodes

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Abstract. Magnetoresistance (MR) in single-walled carbon nanotubes (SWNTs) with different ferromagnetic source and drain electrodes (iron and cobalt) which have different coercivity was studied. Large MR ratio of 20% could be obtained at 8 K, while 1–2% small MR ratio could be observed for the sample with the same ferromagnetic source and drain electrodes of Co. The MR ratio of 20% is very close to the theoretically predicted value of 26% for Co-Fe system.

1. Introduction
Single-walled carbon nanotubes (SWNTs) are the ideal one-dimensional materials, which have been investigated for the viewpoint of nano-device applications [1-5]. Theoretically, one-third SWNTs show metallic characteristics and others semiconducting, which depend on their chirality \((m, n)\) [6]. Charge carriers in metallic SWNTs and multi-walled carbon nanotubes (MWNTs) expect to have a long coherent length, and some experiments indicated the ballistic transport [7, 8]. Using the long coherent length of nanotubes, spin-valve structures have been fabricated with MWNTs or SWNTs [4, 5]. Conventional nanotube spin-valve structures consist of ferromagnetic metal electrode 1, carbon nanotubes and ferromagnetic metal electrode 2, which electrodes 1 and 2 were often the same materials, for example, Co-nanotube-Co or Fe-nanotube-Fe. Magnetoresistance (MR) could be observed in their spin-valve structures, however, the reproducibility of the samples have been one problem. As the average coercivity of the two electrodes is almost the same, their hysteretic MR depends on magnetization fluctuations at the contact point between carbon nanotubes and ferromagnetic electrodes. In order to realize high-quality spin-valve structures with carbon nanotubes, it can be considered that using two different ferromagnetic metals with different coercivity is very promising [9]. In this paper, we report MR in SWNTs with different ferromagnetic source and drain electrodes.

2. Sample preparation and experimental details
Schematic illustration of the samples is shown in Figure 1. Using the conventional photolithography, patterned 1-nm-thick Co layer which works as a chemical catalyst was deposited by the e-beam evaporator onto the Si/SiO\(_{2}\) substrates. Then, the substrate was put into the thermal chemical vapor deposition (CVD) system and a quarts tube was exhausted by an oil-rotary pump until 10\(^{-2}\) Torr, and the furnace was heated up to 800°C with flowing 1,000 sccm Ar gas. SWNTs were grown at 800°C...
and growth time was 10 min. C$_2$H$_5$OH was used as carbon source. Then, 60-nm-thick source and drain electrodes were deposited. In this work, we prepared three kinds of the samples, which were two different ferromagnetic metal contacted SWNT; Co-SWNT-Fe, the same ferromagnetic metal contacted SWNT; Co-SWNT-Co, and the reference sample which was terminated by nonmagnetic metal; Au-SWNT-Au. The distance between source and drain electrode was about 650~700 nm. All Co-SWNT-Co and Co-SWNT-Fe samples had 20-nm-thick gold cap layer for protection against oxidization of cobalt or iron.

Two-terminal MR measurement were carried out in a magnetic field from -600 to 600 mT. The sample was put into a probing system, which can be cooled down to 8 K by He gas cryostat. Field sweep rate was 1.5 mT/sec. Field direction is parallel to the Si substrate and perpendicular to the SWNT. The magnetic field was swept first from -600 to 600 mT, then back to -600 mT.

![Figure 1. Schematic illustration of the SWNT spin-valve structure. The combination of source and drain electrodes were Au-Au, Co-Co and Co-Fe.](image)

### 3. Results and discussion

At first, we measured the reference samples with nonmagnetic terminals; Au-SWNT-Au. Figure 2 shows typical two-terminal resistance as a function of magnetic field at 8 K with excited source-drain voltage ($V_{SD}$) of 1 mV. No hysteretic MR characteristics could be observed.

![Figure 2. Two-terminal MR as a function of magnetic field from Au-SWNT-Au sample. Solid line (dashed line) shows sweep-up (sweep-down) direction.](image)

Results from the same ferromagnetic metals (Co) contacted SWNT is presented in Figure 3. The resistance peak appeared nearby the magnetic field of 0 T. In this sample, about 2% of hysteretic MR
characteristics were observed. These resistance peaks indicate that the contact magnetization had parallel alignment under magnetic field until 0 T, then the magnetization polarity was switched to anti-parallel state. However, the MR ratio was very small (2%) because the average coercivity of the source and drain Co electrodes were almost the same value. MR from two different ferromagnetic metals contacted SWNT (Co-SWNT-Fe) is shown in Figure 4. Large hysteretic resistance peaks could be observed at 8 K. The MR ratio is about 20% in the Co-SWNT-Fe sample.

Figure 3. Two-terminal MR as a function of magnetic field from the same ferromagnetic contacted SWNT sample; Co-SWNT-Co. MR ratio is about 2% and 1% for sweep-up direction and sweep-down, respectively.

Figure 4. MR versus magnetic field of the two different ferromagnetic contacted SWNT sample; Co-SWNT-Fe. MR ratio is about 20% and 15% for sweep-up direction and sweep-down, respectively.

Generally, MR ratio is expressed the equation (1),

$$\frac{\Delta R}{R_p} = \frac{R_{ap} - R_p}{R_p} = \frac{2P_1P_2}{1 + P_1P_2},$$

(1)

where $R_{ap}$ and $R_p$ are resistance in spin anti-parallel state and parallel state, respectively, and $P_1$ and $P_2$ are conduction electrons spin polarizations in the ferromagnetic contacts 1 and 2 [10]. The spin polarization of cobalt and iron is 34% and 44%, respectively [11]. Therefore, $\Delta R/R_p$ of Co-SWNT-Co and Co-SWNT-Fe should be 21% and 26%, respectively. Experimentally, Co-SWNT-Co device showed 2% MR ratio, which was about 10% of theoretical value so that 10% of spin polarized electrons travel through 700 nm without spin-flipping. Since this small MR ratio can be considered to be due to local magnetization fluctuations, it is difficult to control the spin states switching using the same ferromagnetic metal which has the same coercivity. On the other hand, Co-SWNT-Fe sample showed large MR ratio of 20%, which was very close to the theoretical $\Delta R/R_p$ value of 26% for Co-Fe system. This large MR ratio probably caused by different coercivity of source and drain electrodes.

About 30 contacting Co-SWNT-Co and Co-SWNT-Fe samples were measured, however, only one and four of these showed hysteretic MR characteristics, respectively. One problem is the device structures. It can be considered that the 20-nm-thick gold cap layer contact to the SWNT, preventing spin injection. Formation of insulated passivation film at contacting area between ferromagnetic metal and SWNT, for example SiO$_2$ or SiN, is probably useful for SWNT spin-valve structure.

It should be noted that the sweep-down MR peak of Co-SWNT-Fe sample (dashed line in Fig. 4) started at about 200 mT. MR peaks ordinarily start around 0 T like Fig. 3. This shift could be observed other Co-SWNT-Fe samples in greater or lesser degrees. These peak shifts indicate the shift of
magnetization curve. In this case, it can be considered that the cobalt catalyst layer is important. During or after SWNT growth, Co became CoO by oxidization, which has antiferromagnetic characteristic with Néel temperature of around room temperature. Therefore, exchange anisotropy was caused in CoO/Co(Fe) interface and enhancement of coercivity occurred, resulting in appearance of the large MR shift [12].

4. Summary
Different ferromagnetic metals with different coercivity contacted SWNT spin-valve structures were fabricated. Large MR ratio of 20% could be observed in Co-SWNT-Fe sample which was very close to the theoretical value while Co-SWNT-Co showed 2% MR ratio. These results indicate that it is very suitable to fabricate spin-valve structures with SWNTs using two different ferromagnetic metals.

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