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Description	

Fabrication and Characterization of Carbon Nanotube Field-Effect Transistors using Ferromagnetic Electrodes with Different Coercivity

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We have succeeded in fabricating source and drain structures of carbon nanotube field-effect transistors (FETs) by adopting ferromagnetic electrodes with different coercive fields. The electrodes were successfully bridged with single-walled carbon nanotubes (SWNTs) by a direct growth method. We investigated magnetic properties of electrodes and FET characteristics. The magnetic properties of electrodes survived the chemical vapor deposition process at up to 800℃, and were found to be qualitatively preserved even at growth times of 20 and 30 min. In addition, the devices also showed good field-effect modulation in conductivity. This device structure could be applied to carbon nanotube spintronics devices fabricated by a direct growth method.

KEYWORDS: SWNTs, FET, alcohol catalytic chemical vapor deposition, direct growth, shape anisotropy electrodes, coercive field

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1. Introduction

In addition to their high electronic conductivity, carbon nanotubes (CNTs) possess a long spin diffusion length, due to their ideal π-electronic system with small spin-orbit interactions. The long spin diffusion length gives rise to spin-coherent transport, which suggests potential characteristics for spintronics devices. However, details of spin-dependent transport properties of CNTs have not yet been clarified. For the precise control of the charge and spin degrees of freedom, three basic processes, (namely, injection, transport and detection) have to be clarified. Although spin injection to and spin detection from CNTs seem feasible, magnetoresistance (MR) effects of CNTs are mostly observed at low temperature with large variation of MR ratios.[1,2] Naber et al.[3] stated that the MR in a junction of ferromagnetic-electrode/CNT/ferromagnetic-electrode also depends on the properties of the interfaces between ferromagnetic electrodes and CNTs. The reduction and variation of MR ratios can be attributed to poor quality of the CNT-interface contacts, due to contamination of pre-dispersion[4] or post-dispersion[5] medium during the device fabrication process. The method of disposition of CNTs between the ferromagnetic electrodes is one of the most important issues to be established.

Our experimental approach is to directly bridge single-walled carbon nanotubes (SWNTs) between ferromagnetic (Co) electrodes by a direct growth method. This method is effective in reducing the carrier injection barrier height between the electrodes and SWNTs, hence the contact resistance.[6] This could be one of the potential approaches for the enhancement of spin injection and spin detection. In direct growth method, SWNT grows from the Co nanoparticles, which are formed from thin films at high temperatures

during alcohol catalytic chemical vapor deposition (ACCVD) process. Spin-dependent transport is expected to be governed by the magnetic properties of Co nanoparticles to which the SWNT is attached. However, the magnetic property of an individual Co nanoparticle in contact with an SWNT is difficult to measure.[7] The attention of the present work is focused on the fabrication of CNT devices directly from precisely-controlled ferromagnetic electrodes for the realization of spin-valve effect. In spin-valve devices, the resistance is high when the ferromagnetic moments in both electrodes are antiparallel to each other, and low when the ferromagnetic moments are parallel. There are two common methods to realize this effect. One method is to fabricate source and drain electrodes by different ferromagnetic materials with different coercive fields. Another method is to use the same ferromagnetic material, but with source and drain electrodes designed with different aspect ratios, in order to make mutually different coercive fields due to shape anisotropy. The former method is quite difficult to realize because it requires a complicated fabrication procedure. The latter technique is more suitable because of its straightforward fabrication, enabling us to control the coercive field of each electrode. In this communication, we report magnetic properties of the electrodes and their suitability for application to spintronics devices under the growth condition of SWNTs.

2. Experimental method

The geometry of the electrodes fabricated by electron beam (EB) lithography and EB deposition are illustrated in Figure 1. First, 9,000 sets of ferromagnetic electrodes were fabricated on a Si/SiO_2 substrate (5 mm × 5 mm) to enhance the total magnetic moments for the study of the magnetic properties. The widths of source and drain electrodes were

designed to be 0.1 μm and 1.0 μm, respectively, to obtain different coercive fields of electrodes due to the shape anisotropy.[8)] Thickness of electrodes was 100 nm, and thin-film with nominal thickness of 1 nm was attached to an edge of the electrodes by EB deposition method for effective growth of SWNTs. We adopted an angle deposition technique[7)] to form Co thin films at the edge of Co electrodes. In the angle deposition technique, the device was set 8º from normal during Co thin films formation, in order to attach the thin films to the Co electrodes. A double layer of resists, LOL2000 (Rohm and Hass) as the lower layer and ZEP520A-7 (ZEON) as the upper layer, was adopted for EB lithography. The ZEP520A-7 layer was developed by using ZED-N50 (ZEON). Subsequently, the LOL2000 layer was etched by using NMD-W (Tokyo Ohka). This creates an undercut at the lower layer, and allows Co thin films to be attached to the edge of the Co electrodes during the angle deposition. SWNTs were grown at 800 ^{o}C for 10-30 min by ACCVD technique. Detailed procedures regarding SWNT growth were reported previously.[6,7,9)] The magnetic properties of electrodes with and without SWNT growth process were measured by superconducting quantum interference device (SQUID) magnetometer (Quantum Design MPMS) at 300 K and 4.2 K.

Second, for transport measurement studies, we fabricated 12 sets of CNT field-effect transistors (FETs) in a substrate with exactly the same geometry of ferromagnetic electrodes as samples for the magnetic properties measurement. The FETs fabricated in this study had the back-gate configuration. A heavily doped *n*-type silicon wafer with a 400 nm-thick thermally oxidized SiO_2 layer on the surface was used as the substrate (15 mm × 15 mm). The SiO_2 layer and the doped silicon layer of the wafer were used as the gate insulator and gate electrode, respectively. The channel length L was fixed at 1 μm.

Transport properties, including field-effect modulation, were investigated by DC method with semiconductor characterization system at room temperature in vacuum (Dessert TT-prober, Keithley 4200-SCS). The SWNTs grown on the substrate were characterized by scanning electron microscopy (SEM, Hitachi S-4100) and Raman spectroscopy (Tokyo Instruments Nanofinder 30) with a laser excitation energy of 1.96 eV (λ = 632.8 nm). For Raman spectroscopy, the laser spot size was about 2 μm in diameter.

3. Results and discussion

Figures 2 (a)-(h) show magnetic properties of 9,000 sets of source and drain electrodes with and without SWNT growth process, measured at 300 K and 4.2 K. Magnetic properties for non-annealed samples (without SWNT growth process) were measured at both temperatures, for a control experiment. The results at 300 K and 4.2 K are shown in Figure 2 by open circles and open squares, respectively. At both measured temperatures, hysteresis in magnetization curve (M *vs* H) was detected, as usually observed in ferromagnetic materials. In addition, magnetization curves with two clear steps were observed at both temperatures, due to the shape anisotropy of electrodes. These can be attributed to different coercive fields, corresponding to the electrodes with different widths. The small signal for larger coercive field can be attributed to the small volume of the narrow (0.1 μm) electrode, while the larger signal for smaller coercive field corresponds to the wide (1 μm) electrode. The coercive fields for the electrodes with widths of 0.1 μm and 1 μm were estimated to be 250 Oe and 100 Oe, respectively, at 300 K, as shown by open circles in Figure 2 (a). At 4.2 K, hysteresis in magnetization curve was enhanced and the coercive fields increased in comparison with those at 300 K, due to negative thermal coefficient of coercive field, as observed in most ferromagnetic

materials. The coercive fields become 300 Oe and 200 Oe for electrodes with widths of 0.1 μm and 1 μm, respectively, as shown by open squares in Figure 2 (b). It should be noticed that the two steps in the magnetization curve were still observed at 4.2 K.

To study the effects of CVD growth process on the magnetic properties of the electrodes, growth time was varied from 10 to 30 min. The growth temperature was kept the same (800℃) at each growth time for comparison. Figure 2 (c) shows a magnetization curve for electrodes with growth time of 10 min by closed circles: the data for non-annealed sample are also shown by open circles for comparison (data are the same as those in Fig. 2 (a)). The coercive fields for electrodes with widths of 0.1 μm and 1 μm were estimated to be 110 Oe and 70 Oe, respectively, at 300 K. Magnetic switching of narrow and wide electrodes, that is, two steps in the magnetization curve, can clearly be seen at 300 K, even after annealing for 10 min under the growth conditions. Similar behaviour was also observed in samples annealed for 20 and 30 min, as shown in Figure 2 (e) and (g), respectively. Even though the growth time was varied, the coercive fields did not show growth time dependence. On the other hand, the two steps in the magnetization curve became unclear at 4.2 K, although the increase in hysteresis and enhancement of coercive fields were consistent with the result on non-annealed samples. It is well-known that the coercive field strongly depends on the morphology, grain size, etc. of ferromagnetic materials, and it also depends on the surface condition.[10)] Our results show that these factors which govern the coercive fields become inhomogeneous in each electrode through annealing, and at low temperature these become significant, because phenomena are very sensitive in low energy regime. Although the two steps in hysteresis are clear with homogeneous film at 4.2 K, the two steps in hysteresis became

unclear after CNTs growth. Even though the averaged magnetization curves of 9,000 sets of electrodes did not show two steps in hysteresis due to surface inconsistencies as a result of annealing, we can expect to see a local magnetization curve which still shows two step hysteresis.

Figure 3 (a) shows an SEM image of the area between the electrodes after ACCVD for 20 min. The left electrode has width of 1 μm, and the right electrode has width of 0.1 μm. Fine structures between the electrodes were observed, and it was found that some of the CNTs bridged the two electrodes. These fine structures were confirmed to be SWNT based on Raman spectra analysis. Although we adopted Mo support layer for the stabilization of electrodes in previous works,[6,7] SWNTs also successfully bridged the electrodes using only ferromagnetic materials (Co). Figure 3 (b) shows a Raman spectrum obtained from the channel region between the source and drain electrodes. The Raman spectrum exhibits radial breathing mode (RBM) peak at 162 cm^{-1} and G-band peak at 1590 cm^{-1}. The strong peaks at 300, 520, and 950 cm^{-1} originate from the Si substrate. Although a peak exists at D-band region (1320 cm^{-1}), RBM and G-band peaks are dominant, and they match those in typical Raman spectra of CNTs.[11] I_G/I_D ratio was calculated to be nearly 5. This value is comparatively small[7] due to the formation of unwanted amorphous carbon on the surface of electrodes. Note that Raman spectrum was measured in the channel region where the channel length L was fixed at 1 μm. Since the laser spot size is 2 μm, which is larger than the channel region, the peak information is not only from the channel region, but also from the neighboring electrode region. Therefore, a rather high D-band peak was observed. For reference, a study on SWNT growth from Co catalyst thin films on a wide Si/SiO_2 surface was also performed to

measure the quality of the CNTs, and the result showed high purity of SWNTs with a very low D-band peak.[12)]

Figure 4 shows output and transfer characteristics of a CNTFET device (growth time, 20 min) at 300 K. In this measurement, the left electrode of the device in Figure 1 was grounded as the source electrode. Field-effect modulation in conductivity (which shows *p*-type operation) was observed. We fabricated 12 devices in one substrate, where more than half of them showed FET operation. The on-off ratio and threshold voltage for data measured at 300 K (with V_{DS} = -10 V) from the transfer curve were 1.2×10^2 and -1.7 V, respectively. Even though the electrodes were only formed with ferromagnetic material and with different aspect ratios, the device characteristics are comparable to our previous report on CNTFETs with symmetric electrodes.[6)]

4. Conclusions

We have fabricated CNTFETs by means of direct growth method for bridging SWNTs between ferromagnetic electrodes with the shape anisotropy. These electrodes consist of only ferromagnetic (Co) electrodes, which could act as spin injector and spin detector through the SWNTs. The device operation was consistent with CNTFETs with symmetric electrodes. The magnetic properties of electrodes withstood the CVD process at up to 800 ^{o}C through the growth of SWNTs, and can withstand growth time up to 30 min. The method presented in this study will contribute to the possibility of fabrication of single-walled carbon nanotube spin-valve devices by direct growth method.

Acknowledgement

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Figure Captions

Fig. 1. (Color online) Top view (top) and side view (bottom) of Co electrode geometry with different aspect ratios. Gray and dark blue areas correspond to Co thin-films formed by normal and angled deposition, respectively. Magnetic field was applied to the electrodes' long-axis direction for magnetic property measurement by SQUID.

Fig. 2. (Color online) Magnetization curves of 9,000 sets of electrodes without annealing measured at 300 K (a) and 4.2 K (b), annealed for 10 min and measured at 300 K (c) and 4.2 K (d), annealed for 20 min and measured at 300 K (e) and 4.2 K (f), and annealed for 30 min and measured at 300 K (g) and 4.2 K (h). Data of non- annealed electrodes shown in (a) and (b) are also plotted, respectively, in (c), (e), (g) by open circles, and (d), (f), (h) by open squares for comparison.

Fig. 3. (a) An SEM image of the area between the electrodes showing SWNTs bridging the electrodes (growth time, 20 min). (b) A Raman spectrum measured at the channel region between source and drain electrodes.

Fig. 4. (a) (Color online) Output and (b) transfer characteristics of the device at 300 K (growth time, 20 min). In figure 4 (b), V_{DS} = -10 V. Arrows indicate forward and backward sweep direction of the gate voltage.

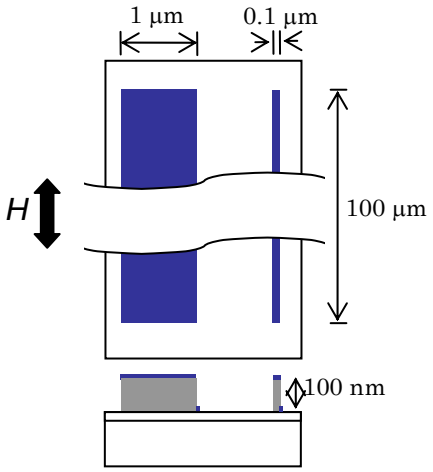

Fig. 1

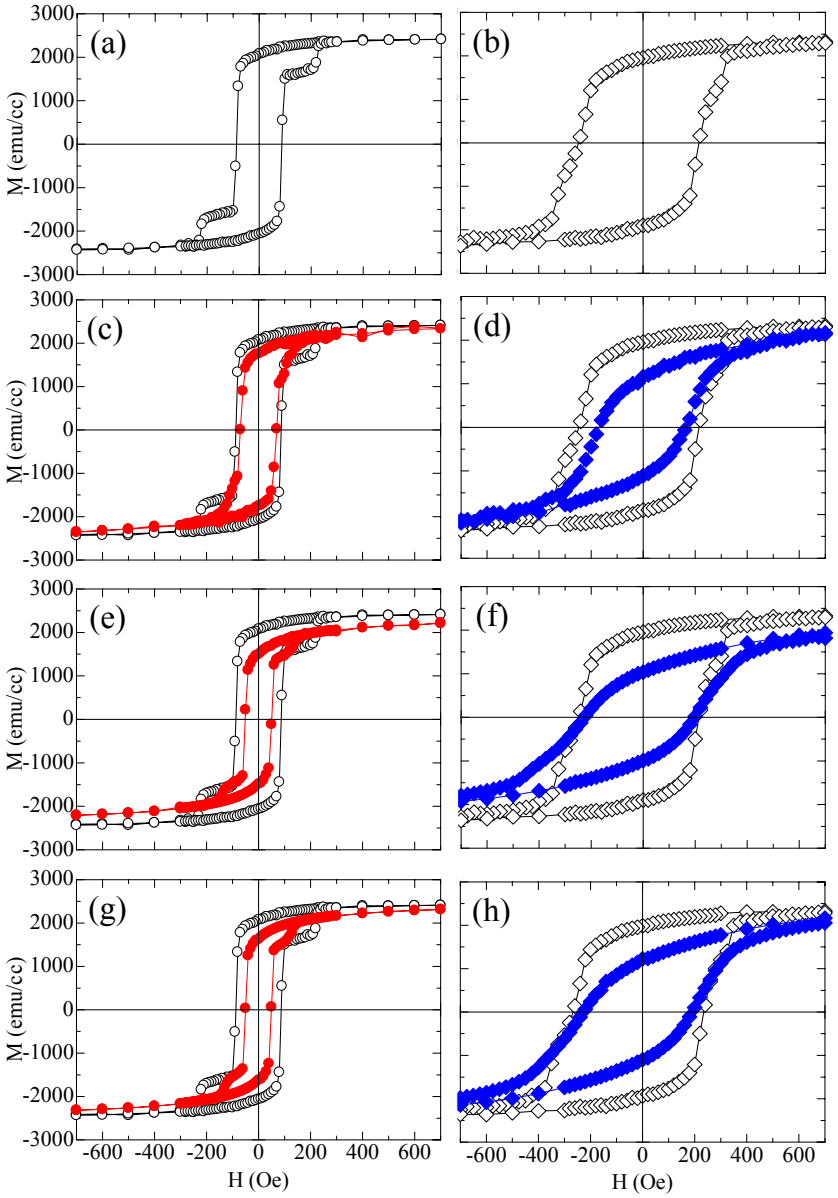
(a)
(b)
(c)
(d)
(e)
(f)
(g)
(h)
M (emu/cc)
H (Oe)
3000
2000
1000
0
-1000
-2000
-3000
-600
-400
-200
0
200
400
600

Fig. 2

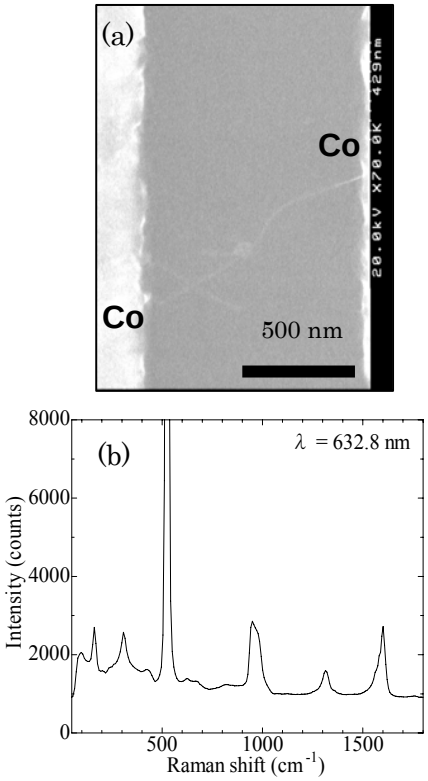
(a)
Co
Co
500 nm
20.0kV X70.0K 429nm
(b)
λ = 632.8 nm
Intensity (counts)
8000
6000
4000
2000
0
500
1000
1500
Raman shift (cm-1)

Fig. 3

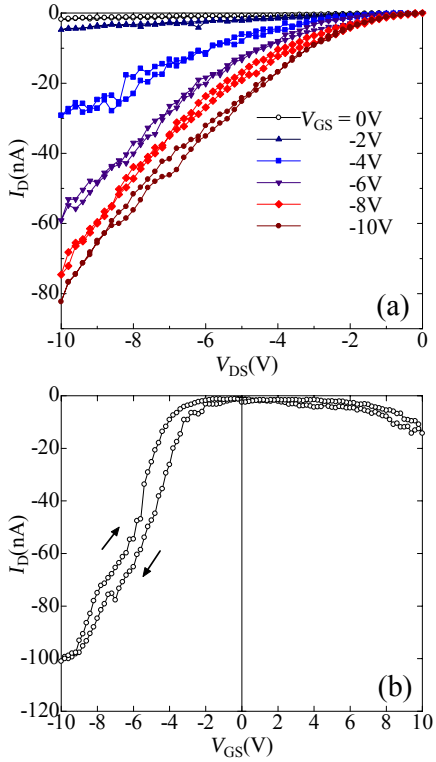
0
-20
-40
-60
-80
I_D(nA)
V_{GS} = 0V
-2V
-4V
-6V
-8V
-10V
(a)
-10
-8
-6
-4
-2
0
V_{DS}(V)
0
-20
-40
-60
-80
-100
-120
I_D(nA)
(b)
-10
-8
-6
-4
-2
0
2
4
6
8
10
V_{GS}(V)

Fig. 4