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Spin-polarized transport in carbon nanotubes

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We present transport measurements of ferromagnetically contacted carbon nanotubes. In both single- and multi-walled nanotube devices, a spin valve effect is observed due to spin-polarized transport. In one single-walled nanotube device, the spin-valve effect is suppressed as the influence of Coulomb charging is observed at around 10 K. To help understand the interplay between the Coulomb charging and the spin-polarized transport we investigated the temperature dependence of the carbon nanotube magnetoresistance.

Key words: carbon nanotube, spin-polarized transport.

1. Introduction

The carbon nanotube is a promising candidate as a component in nanoscale molecular electronic devices [1–9]. It acts as a molecular wire, with conducting properties that are far better than a metal wire of similar dimensions. Because the carbon nanotube is a stable, self-assembled structure, its width is uniform, and it is relatively defect-free [1–3]. In addition, metallic nanotubes show extremely high conductivity, and a long mean-free path. Ballistic transport has been observed in nanotubes even at room temperature [4]. The spin scattering length in carbon nanotubes is also expected to be quite long, opening the possibility of creating a carbon nanotube magnetoelectronic device [5, 6]. In this report, we explore this possibility and present measurements of electron spin transport in carbon nanotubes.

2. Sample preparation and experimental details

We disperse nanotubes onto a SiO_2/Si substrate, and map out the position of nanotubes with respect to Pt/Au alignment marks on the substrate using a scanning electron microscope. We use multi-walled carbon nanotubes (MWNTs) or single-walled carbon nanotubes (SWNTs). Contact patterns are defined so as to overlap the mapped nanotube using electron-beam lithography, after which 65 nm of Co is deposited by thermal evaporation at a pressure of 4×10^{-7} Torr. The resulting polycrystalline Co film behaves ferromagnetically and has a room-temperature resistivity of approximately 22 $\mu\Omega$ cm. After lift-off, the ferromagnetic leads are connected to nonferromagnetic bond-pads.

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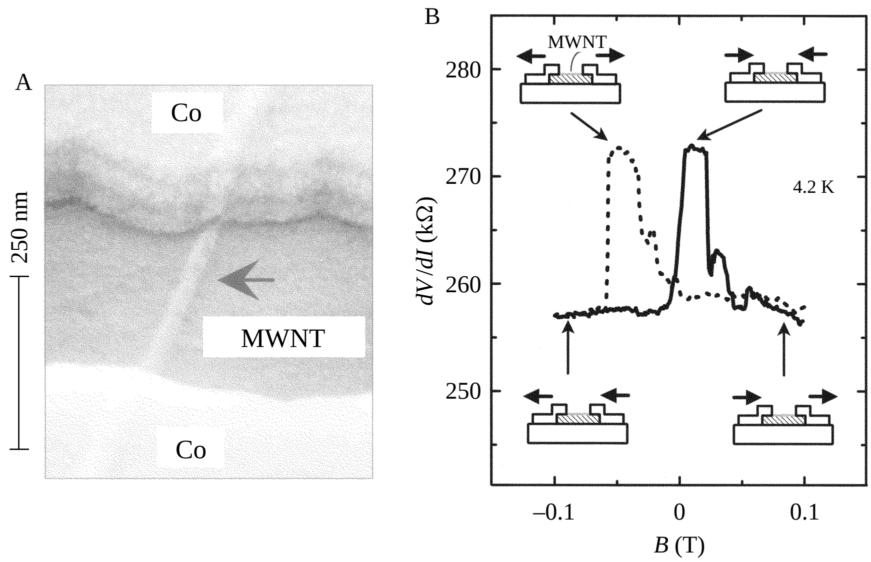

Fig. 1. A, Micrograph of a spin-valve device consisting a single multi-walled carbon nanotube (MWNT) electrically contacted by ferromagnetic Co. The Co contacts lie on top of the MWNT, and the conducting channel is approximately 250 nm in length. B, Two-terminal differential resistance as a function of magnetic field. The magnetic field is directed parallel to the substrate, and the temperature is 4.2 K. The solid (dashed) trace corresponds to the positive (negative) sweep direction. The magnetization direction of the left and right contacts is represented by the direction of the arrows in the figure. The percentage difference $\Delta R/R_0$ between the tunnel resistance in the parallel and the antiparallel states is approximately 6%.

3. Results and discussion

3.1. Multi-walled carbon nanotube

Crude MWNTs synthesized from graphite rods by the arc discharge evaporation method under He atmosphere [2] are used. This ensures that the MWNTs contain no trace of magnetic impurities. The MWNTs are typically 10–50 nm in diameter and a micron or more in length. Figure 1A is an electron micrograph showing the junction region of a completed device.

Magnetoresistance measurements are performed in a 4.2 K bath cryostat with the B-field directed in the plane of the substrate. The two-terminal resistance is measured using an ac lock-in technique with an excitation voltage of 100 μV. Figure 1B shows the two-terminal differential resistance of a Co-contacted nanotube as a function of magnetic field. The field is swept first from -100 mT to 100 mT (solid line) and then back to -100 mT (dashed line). A resistance peak appears as the magnetic field moves through 0 T. There is also a large hysteresis in the peak position ($\pm$50 mT) between positive and negative sweep directions, indicating the probable influence of the contact magnetization.

Similar hysteretic magnetoresistance is observed in magnetic tunnel junctions (MTJs), where it has been attributed to spin-polarized electron tunneling [10–15]. The MTJs consist of two ferromagnetic contacts separated by a thin oxide layer. The conduction electrons within the ferromagnetic contacts have a preferred spin direction, which is determined by the local magnetization. This causes the formation of majority and minority spin conduction bands with different densities of states at the Fermi energy. In the absence of spin-scattering, the resistance across the tunnel barrier is dependent on the relative alignment of the magnetization of the two contacts. In the anti-parallel state the majority spin states are out of alignment and the junction resistance is higher than in the parallel state in which the majority spin states are aligned. For the nanotube

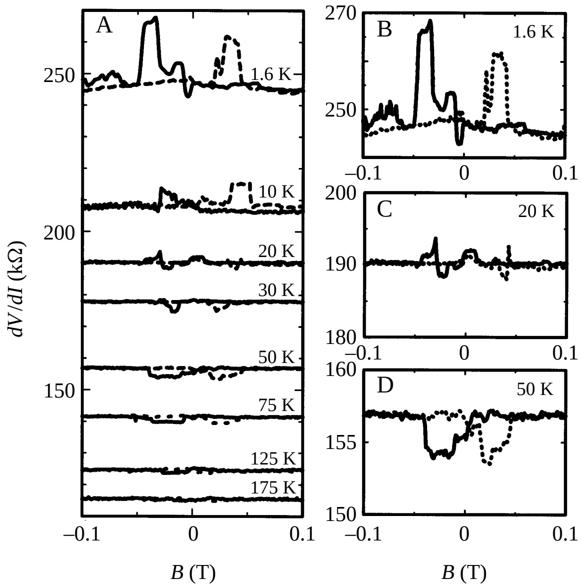

Fig. 2. A, Temperature dependence of the two-terminal differential resistance as a function of magnetic field. B, At low temperatures, the positive spin-valve signal comes out. C, The signal almost disappears at 20 K and D, flips to negative at higher temperature. The spin-valve signal eventually disappears at 175 K.

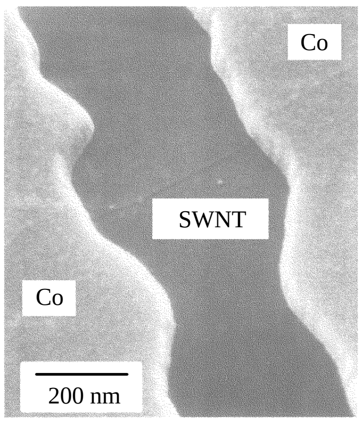

Fig. 3. Micrograph of a single bundle of single-walled carbon nanotubes (SWNTs) electrically contacted by ferromagnetic Co. The Co contacts lie on top of the SWNT, and the conducting channel is approximately 250 nm in length. A side-gate is located at 5 μm apart from the nanotubes.

devices, the contact magnetizations align parallel with the magnetic field at $B = 100$ mT and -100 mT (Fig. 1B). As we sweep B through 0 T, the magnetization polarity switches. The observed peak suggests that the contact magnetizations switch separately and become misaligned as the field is swept. For a MTJ, misalignment occurs because different ferromagnetic contact materials are used, with different coercivities—the magnetizations are misaligned when B lies between the coercive fields of the two contacts. This does not explain the misalignment in the nanotube device. The misalignment may be caused by magnetization fluctuations that occur locally, on the scale of the nanotube diameter (30 nm). The average Co domain size

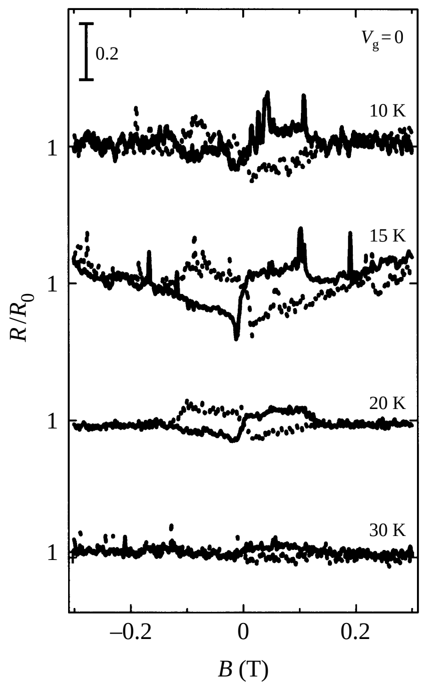

Fig. 4. Two-terminal differential resistance normalized by the zero-field resistance as a function of magnetic field at various temperatures. When the magnetic field is applied parallel to the contact plane, the magnetoresistance has a peak associated with the spin-valve effect between the two ferromagnetic contacts. The resistances are normalized by each zero-field resistance; 2610 kΩ for 10 K, 290 kΩ for 15 K, 175 kΩ for 20 K, and 130 kΩ for 30 K. Solid lines and dotted lines are taken in different field-scan directions. The gate voltage is 0.

(50 nm) [16] is on the order of the width of the nanotube so that the nanotube contacts only a small number of magnetic domains. The coercivity of each domain varies, and depends on its geometry and the local energy conditions.

The sample-to-sample variations observed are probably due to inherent random variations in the surface condition over the small nanotube contact area. Previous experiments on nonmagnetically contacted nanotubes have observed large variations in the contact resistance [7]. Also, in the ferromagnetically contacted samples it is impossible to control the particular domain structure in contact with the nanotube.

The spin-injection picture for the nanotube magnetoresistance requires that a sufficiently small amount of spin scattering occurs both within the nanotube, and at the interfaces between the nanotube and the contacts. Following Julliere's model based on the magnetic tunnel junction [13], the spin-scattering length $l_s \sim 130$ nm for our best result. Although fairly long, this is probably an underestimation. The spin-polarization near the ferromagnet/nanotube interface will depend on the interface quality.

In the large variations of the domain walls in the polycrystalline Co film, we found a device which shows striking temperature dependence (Fig. 2). As shown in Fig. 2B, the positive magnetoresistance flips to negative at about 20 K while the resistance peak generally becomes smaller as the temperature increases. The polarity-flip was repeatable even after thermal cycles. We suppose that the flip may be attributed to the reconstruction of the domain contributing to the transport in one of the ferromagnetic electrodes. The domain walls pinned at low temperature could be released at higher temperatures to achieve more stable domain walls in the electrodes. Another interpretation uses a model of the two dominant domains in one side of the contacts—one domain has aligned magnetization with the magnetic field and the other is misaligned to the magnetization in the first contact—the two have different temperature dependences. For this model, the

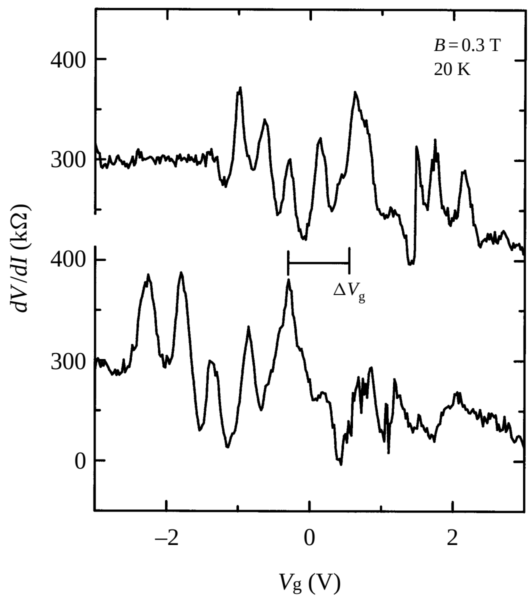

Fig. 5. Gate–voltage characteristics of the ferromagnetically contacted SWNTs with a magnetic field of 0.3 T at 20 K. Upper (lower) trace shows the part of a scan from +4 to −4 V (from −4 to +4 V). The resistance shows oscillations. ΔV_g shows peak shift, which depends on the voltage sweep direction. The value of ΔV_g depends on the applied magnetic field.

domain with misaligned magnetization under the magnetic field survives at higher temperature than the aligned magnetization.

3.2. Single-walled carbon nanotube

For the SWNTs, the transport is more complicated. In the SWNT two-terminal device with a metal contact, single-electron tunneling was observed [8, 9].

We use bundles of SWNTs (less than 10 nm in bundle diameter) synthesized by the pulsed laser vaporization of a metal/carbon target [3]. The typical length of a bundle is 1–20 μm. The polycrystalline Co contacts are formed on the SWNTs in a similar manner to that for the MWNTs. The contact separation is approximately 250 nm (Fig. 3). In the Co-SWNTs device, the spin-valve effect was observed. Figure 4 shows two-terminal differential resistance as a function of magnetic field at various temperatures. In general, the signal of the spin-valve effect becomes more pronounced at lower temperature. In the Co-SWNTs device, the magnetoresistance change, at first, becomes larger nearer to 15 K. At lower temperature, however, the signal was suppressed, and the conductance oscillations, which might be caused by the Coulomb blockade effect, become pronounced in the gate–voltage characteristics. This may indicate that spin polarity of the conduction current is randomized during tunneling through the Coulomb island.

In order to see the co-existing state with the spin coherence and the Coulomb blockade effect, we measured the gate–voltage characteristics at 20 K under the magnetic field of 0.3 T. The resistance oscillates in the gate–voltage characteristics (Fig. 5). The observed peak is hysteretic in gate–voltage scan; the shift of ΔV_g is about 1 V. The reproducible hysteresis with the same oscillations period is observed while we keep the temperature and the applied magnetic field constant. The ΔV_g is largest near the zero-field, and saturated under the higher magnetic fields. Thus, this might be not the conventional current shift due to electron traps near the single electron transistor. We found similar hysteretic gate–voltage characteristics in other device.

The result is appears to be peculiar to carbon SWNTs. Interpretation of this result, however, will require a further investigation.

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References

[1] R. Saito, G. Dresselhaus, and M. S. Dresselhaus, *Physical Properties of Carbon Nanotubes* (Imperial College Press, Singapore, 1998).
[2] T. W. Ebbesen and P. M. Ajayan, Nature **358**, 220 (1992).
[3] A. Hess *et al*., Science **273**, 483 (1996).
[4] S. Frank *et al*., Science **280**, 1744 (1998).
[5] K. Tsukagoshi, B. W. Alphenaar, and H. Ago, Nature **401**, 572 (1999).
[6] B. W. Alphenaar, K. Tsukagoshi, and H. Ago, Physica E **6**, 848 (2000).
[7] A. Bachtold *et al*., Nature **397**, 673 (1999); C. Shöenberger *et al*., Appl. Phys. **A69**, 283 (1999).
[8] S. J. Tans *et al*., Nature **394**, 761 (1998).
[9] M. Bockrath *et al*., Science **275**, 1922 (1997).
[10] P. M. Tedrow and R. Meservey, Phys. Rev. Lett. **26**, 192 (1971).
[11] A. G. Aronov, Pis'ma Zh. Eksp. Teor. Fiz. **24**, 37 (1976) [JETP Lett. **24**, 32 (1976)].
[12] M. Johnson and R. H. Silsbee, Phys. Rev. Lett. **55**, 1790 (1985).
[13] M. Julliere, Phys. Lett. **54A**, 225 (1975).
[14] J. S. Moodera *et al*., Phys. Rev. Lett. **74**, 3273 (1995).
[15] T. Miyazaki and N. Tezuka, J. Magn. Magn. Mater. **139**, L231 (1995).
[16] U. Rüdiger *et al*., Phys. Rev. **B59**, 11914 (1999).