Spin Transport in Nanotubes and Novel Barriers for Magnetic Tunnel Junctions — Paolo Freitas, Chairman

Spin transport in nanotubes (invited)

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We study electron spin transport through carbon nanotubes contacted by ferromagnetic electrodes. The resistance of a ferromagnetically contacted multiwalled nanotube switches hysteretically as a function of applied magnetic field, with a maximum resistance change of 9% at 4.2 K. Magnetoresistance measurements of carbon nanotubes having one cobalt contact and one platinum/gold contact, however, show no switching. In addition, we present calculations of the magnetoresistance ratio for the ferromagnetic nanotube device, and predict that a resistance change of 25% is possible. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1359220]

In spin electronics, the electron spin is used, either in conjunction with or independent of the electron charge, to store and transfer information.[1] This involves: (1) creation of a spin-polarized state to represent a bit of information, (2) transfer of the spin polarization without loss of information, and (3) detection of the spin state and read-out of the stored information. Ferromagnets are a natural source of spin-polarized electrons and can act as either spin injectors or detectors. Spin-polarized electrons can be injected from a ferromagnet into a nonferromagnetic material or through an oxide tunnel barrier. The electron scattering rate at any subsequent nonferromagnetic/ferromagnetic interface depends on the spin polarity. By varying the nonferromagnetic layer composition and the measurement configuration, a variety of successful spin-electronic devices have been created, including magnetic tunnel junctions, spin valves, and giant magnetoresistance devices. In these well-known spin-electronic structures, the nonferromagnetic layer is relatively thin, so that electron spin is transferred only over a short distance (on the order of 2 nm). Extending spin transmission over longer distances and allowing for the possibility of spin manipulation during the transfer process could enhance the role of the electron spin. The use of metals and semiconductors, with spin scattering lengths of 100 μm or more has been suggested for three-terminal spin-electronic devices.[2] However, momentum scattering lengths in metals and semiconductors are much shorter than spin scattering lengths, resulting in diffusive electron transport, and small spin-mediated resistance changes.[3,4]

The carbon nanotube is a recently discovered material system[5] whose novel properties offer intriguing possibilities for spin injection and spin electronics. Carbon nanotubes consist of sheets of graphene wrapped in seamless cylinders. Single walled nanotubes (SWNTs) consist of a single graphene sheet that forms a cylinder 1–3 nm in diameter, while multiwalled nanotubes (MWNTs) are composed of a number of cylinders arranged concentrically in an onion-like fashion, with a maximum diameter of approximately 80 nm. Depending on the chirality (wrapping angle) of the single walled nanotube and its diameter, it can behave as either a semiconductor or a metal. SWNTs have two one-dimensional conducting channels at the Fermi energy, leading to a total conductance of $4e^2/h$. There are a number of reasons why carbon nanotubes should be considered as a possible basis for spin-electronic devices. First, the electron and phase scattering lengths are extremely long, so that the nanotube should behave as a ballistic conductor and allow for larger spin mediated resistance changes than in the diffusive case. Second, spin flip scattering is expected to be particularly small in carbon nanotubes, since carbon is such a light atom. Finally, the carbon nanotube is expected to behave as a ''Lüttinger liquid''[6] and recent theory[7] suggests that spin–charge separation should be observable in a ferro/nanotube/ferro device and produce sawtooth-like oscillations in the current/voltage characteristics.

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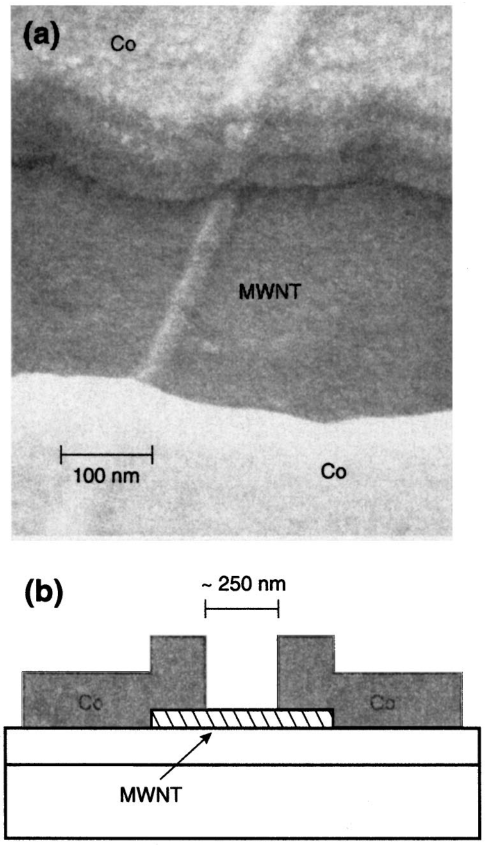

FIG. 1. (a) Scanning electron microscope image of a Co contacted MWNT. The diameter of the nanotube is 30 nm and the conducting channel length is 250 nm. (b) Schematic diagram of the device. The substrate is a semi-insulating Si wafer covered by 200 nm thick SiO_2.

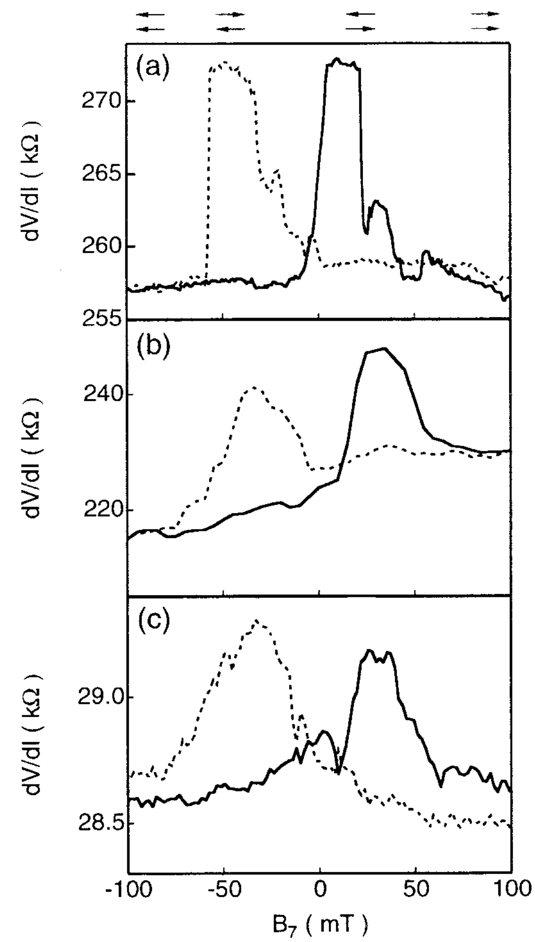

FIG. 2. Two-terminal differential resistance as a function of magnetic field for three different Co contacted MWNT devices. The dashed (solid) line shows the down (up) sweep direction. The magnetic field is directed parallel to the substrate, perpendicular to the current path, and the temperature is 4.2 K.

In this article we will describe our results on ferromagnetically contacted carbon nanotubes. We use ferromagnetic contacts to inject and detect spin-polarized electrons into a multiwalled carbon nanotube. As the alignment of the magnetizations within a pair of contacts switches from parallel to antiparallel, the nanotube resistance switches from a low to a high resistance state, with a maximum resistance change of 9% at 4.2 K. This provides evidence that electron spin can be injected from a ferromagnetic contact into a carbon nanotube and remain polarized over distances of at least 260 nm. This interpretation is complicated by the possible influence of the stray field from the ferromagnetic contacts on the nanotube magnetoresistance. However, stray field calculations, and measurements of single-ferromagnetically contacted devices, suggest that spin injection is in fact the dominant effect. Finally, we present the results of calculations of the transport through a ferromagnetically contacted SWNT using a two-band nonparabolic dispersion relation. In contrast to the results of diffusive models, this ballistic model predicts large spin-mediated resistance changes.

We have performed magnetoresistance measurements of ferromagnetically contacted MWNTs and observed evidence for spin injection.[8] Figure 1 shows the geometry of our ferromagnetically contacted nanotube devices. We use crude MWNTs synthesized from graphite rods by the arc discharge method under a helium atmosphere. This ensures that the MWNTs contain no trace of magnetic catalysts. The MWNTs are extracted into suspension in dichloroethane using a short ultrasonic treatment and then dispersed and dried onto a SiO_2/Si substrate. We map out the MWNTs with respect to Pt/Au alignment marks on the substrate using a scanning electron microscope. The MWNTs are typically 10–40 nm in diameter and 1–10 microns in length. Contact patterns are defined using electron beam lithography, after which a ferromagnetic film (typically cobalt) 60–100 nm thick is deposited by thermal evaporation. After lift-off, nonferromagnetic Au/Ti leads and bond pads are connected to the ferromagnetic contacts. Figure 1(a) is an electron micrograph showing the junction region of a completed device. We have fabricated and characterized more than 100 of these devices. Among these, the room temperature resistance varies between 8 and 250 kΩ. When cooled to 4.2 K, the resistance typically increases by a factor of 2 to 3, in agreement with results for nonferromagnetically contacted MWNTs from the literature.[9]

Magnetoresistance measurements are performed in a helium bath cryostat with the magnetic field from a superconducting magnet directed in the plane of the substrate. The two-terminal resistance is measured using an ac lock-in tech-

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nique with an excitation voltage of 100 μV. Figure 2 shows the two-terminal resistance of three different Co-contacted nanotubes as a function of magnetic field. The field is first swept from -100 to 100 mT (solid line) and then back to -100 mT (dashed line). In each trace a resistance peak appears as the magnetic field moves through 0 T. There is a large hysteresis in the peak position between positive and negative sweep directions, indicating the probable influence of the contact magnetizations.

The contact magnetizations align parallel with the magnetic field at $B \cong \pm 100\,\mathrm{mT}$. As we sweep B through 0 T, the magnetization polarity switches. The observed peaks in the nanotube resistance suggest that the contact magnetizations switch separately and become misaligned as the field is swept. In the antiparallel state, the majority spin states of the ferromagnets are out of alignment and the device resistance is higher than in the parallel state in which the majority states are aligned. The magnetization misalignment may be caused by fluctuations that occur locally on the scale of the nanotube diameter. The coercivity of each domain varies and depends on its geometry and local energy conditions. The small switches in the resistance, seen most clearly in Fig. 2(a), provide additional evidence for local domain magnetization fluctuations.

Our results imply that spin injected from the ferromagnetic contact travels coherently through the nanotube and directly influences the resistance. We can make a rough estimate for the minimum required spin scattering length in the nanotube using Julliere's model[10] for spin injection across a tunnel barrier. The difference between the tunnel resistance in the parallel (R_p) and antiparallel (R_a) states is given by

$$\Delta R/R_a = (R_a - R_p)/R_a = 2P_1P_2/(1+P_1P_2). \quad (1)$$

Here P_1 and P_2 are the percentage of conduction electrons polarized in the majority spin band in the two ferromagnetic contacts. For Co, the polarization has been determined to be 34%,[11] giving a maximum resistance change of 21%. In our best case, $\Delta R/R_a$ reaches a maximum value of 9%, suggesting that some percentage of the spin-polarized electrons scatters either within the nanotubes or at the ferromagnet/nanotube interface. If we assume that the spin polarization reduces as $\exp(-l/l_s)$ within the nanotube, this gives a minimum spin scattering length of 262 nm. Although fairly long, this is probably an underestimation. The spin polarization near the ferromagnetic/nanotube interface will depend on the interface quality and could be appreciably lower than 34%. Also, we do not take into account spin scattering at the ferromagnetic/nanotube interface. A model developed specifically for nanotubes[12] is needed to more accurately describe our results.

The experiments described provide evidence for spin coherent transport in carbon nanotubes, and open up the possibility of developing a spin-electronic carbon nanotube device. However, because the stray fields of the ferromagnetic contacts can also influence the resistance of the MWNTs, the role of the spin in determining the resistance of the nanotube devices needs to be confirmed. The resistance of a MWNT measured with nonferromagnetic contacts typically decreases by about 1% as the applied field increases from 0 to 0.1 T.

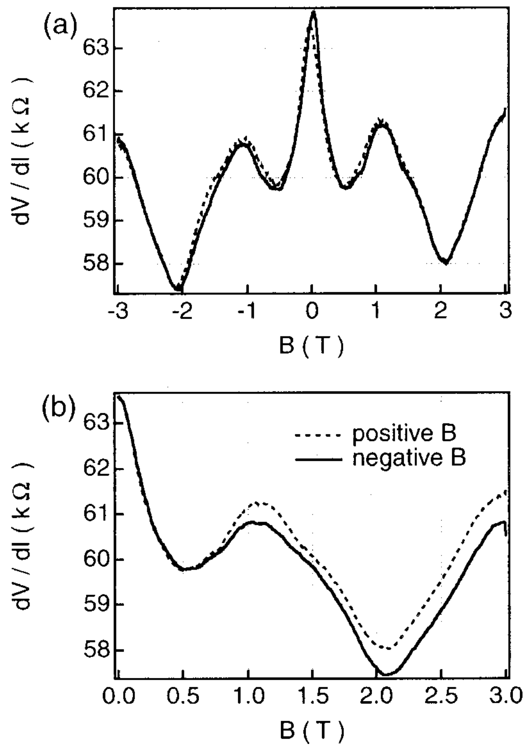

FIG. 3. (a) Two-terminal differential resistance as a function of magnetic field for a MWNT having one Co contact and one Pt/Au contact. The dashed (solid) line shows the down (up) sweep direction. The magnetic field is directed parallel to the substrate, perpendicular to the current path, and the temperature is 4.2 K. (b) Average of the differential resistance measured in the positive (dashed line) and negative (solid line) field directions. Note that the magnitude of the resistance differs between the two field polarities but the magnetoresistance peak positions remain fixed.

This is thought to be mostly due to the influence of magnetic flux on the phase coherent transport pathways in the nanotube.[9] Therefore some fraction of the resistance change that we observe might be due to the changing influence of stray fields from the contacts. To determine the possible magnitude of this effect, we calculated the maximum magnetic field produced in a 100 nm gap separating two semi-infinite 50 nm thick cobalt layers. The contact magnetization was taken to equal its saturation value ($M_s = 1400\,\mathrm{kA\,m^{-1}}$). For the configuration of the measurements in Fig. 2, in which the applied field is in the contact plane but perpendicular to the current direction, the maximum change in field between the parallel and antiparallel states is approximately 0.03 T. This should produce a resistance change of only 0.3%, which is much smaller than the 9% change that we observe.

Another possibility is that fluctuations in the domain configuration of the ferromagnetic contacts produce a much larger local magnetic field variation than is indicated by the calculation for the homogeneous contact. It is difficult to estimate the magnitude of this effect, however, we can experimentally test this hypothesis by performing magnetoresistance measurements on devices having only a single ferromagnetic contact. Resistance changes due to the local stray field should be observed in a MWNT having only a single ferromagnetic contact, while resistance changes due to spin injection require two ferromagnetic contacts (a spin injector

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and detector). To test this prediction, we fabricated five MWNT devices contacted by one ferromagnetic contact (Co) and one nonferromagnetic contact (Pt/Au). Figure 3(a) shows results from a typical device. A small hysteresis in the magnetoresistance is observed around $B=0$, presumably produced by the stray field from the contact. However, no hysteretic switching similar to that in Fig. 2 is observed in this or any of the single ferromagnetic contact devices. Although this evidence is suggestive, there is of course still some possibility that if more single ferromagnetic contact devices were made, one of them would show switching due to changes in the domain configuration. This is because of the inherently large sample-to-sample variations in carbon nanotube device characteristics.[8]

An interesting aspect of the data is that the magnetoresistance is asymmetric with magnetic field, up to 3 T. A similar asymmetry is observed in the majority of our double magnetic contact devices.[13] The exact reason for this asymmetry is still not understood. Nevertheless, it does imply that there is a difference in the ferromagnetic domain configuration between the positive and negative field directions and that this difference survives up to surprisingly high fields. (We have done additional experiments showing that the asymmetry survives up to 6 T.) Note that it is not obvious that the asymmetry is caused by a change in the stray field because the magnetoresistance peak positions (which depend on the applied field) are independent of the magnetic field direction.

For the development of a spin-electronic carbon nanotube device, it would be useful to determine the maximum possible spin-mediated resistance change, and the conditions under which this maximum can be observed. Recently, the diffusive model for ferromagnetic/nonferromagnetic/ferromagnetic multilayer structures[14] has been reinvestigated for semiconductor spin-injection devices,[3,4] and the spin-mediated resistance change predicted to be less than 1%. This prediction is rather discouraging, but fortunately for our study, it is not particularly relevant for the ferromagnetic/nanotube device. In a carbon nanotube the electron and phase scattering lengths are extremely long, on the order of 10 μm or more at 4.2 K.[5] This means that the carbon nanotube is a ballistic conductor, and electron wave functions should be considered as coherent over the length of the nanotube. Recently, it has also been suggested that the spin-mediated resistance change in a ballistic system should be small, since the small number of one-dimensional channels in the ballistic conductor will be filled approximately equally by both spin polarities.[4] While this might be true if the potential changes adiabatically at the contact interface (as for a quantum point contact[15]), this reasoning does not apply for a nonadaibatic contact potential, such as a tunnel barrier. It is expected that the change in resistance for this ballistic system should be closer to that of a magnetic tunnel junction, where electron coherence is also important. A newly published theory[12] developed to describe the ferro/nanotube/ferro devices predicts that a spin-mediated resistance change should be observable, and that $\Delta R/R_a$ can be as high as 20%.

We have performed preliminary calculations of the transmission through a ferromagnetic/SWNT/ferromagnetic structure, taking into account the coherence of the electron wave functions within the nanotube. We use a two-band model for the conduction electrons in the ferromagnets to describe the d and s-p bands that are partially filled in Fe, Co, or Ni. The d band is characterized by a nonparabolic dispersion relation and is assumed to be completely spin polarized, while the s-p band is free-electron-like, with randomly oriented spin. We assume a (5,5) armchair SWNT, in which the dispersion relation consists of a pair of bands that meet degenerately at the Fermi energy, and allow for metallic conduction.[5] The contacts between the ferromagnet and the nanotube are modeled by delta function potential tunnel barriers. Figure 4(a) shows the one-dimensional transmission coefficients for the parallel ($T_{\uparrow\uparrow}$) and the antiparallel ($T_{\uparrow\downarrow}$) configurations as a function of electron energy near the Fermi energy. Sharp peaks are observed, indicating resonances due to partial backscattering at the carbon nanotube/ferromagnet interfaces.

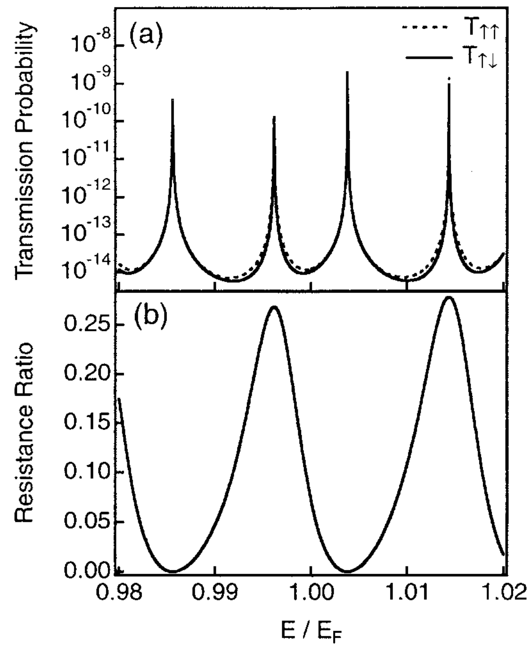

FIG. 4. (a) Transmission probability for the parallel ($T_{\uparrow\uparrow}$) and antiparallel ($T_{\uparrow\downarrow}$) configurations calculated for a ferromagnet/SWNT/ferromagnet device using the ballistic model described in the text. (b) The resistance ratio $\delta R=(T_{\uparrow\downarrow}-T_{\uparrow\uparrow})/(T_{\uparrow\downarrow}+T_{\uparrow\uparrow})$ calculated using the results in (a).

If we assume that the current is proportional to the transmission probability (i.e., the zero temperature, linear response result), the resistance ratio can be defined as $\delta R=(T_{\uparrow\downarrow}-T_{\uparrow\uparrow})/(T_{\uparrow\downarrow}+T_{\uparrow\uparrow})$. (More realistically, we should integrate over the lateral density of states perpendicular to the tunneling direction to determine the current. This calculation is now in progress.) In Fig. 4(b), the resistance ratio δR is plotted as a function of energy using the results from Fig. 4(a). The exact value for δR is dependent on a number of parameters, not all of which are known experimentally. These include the precise form of the ferromagnet dispersion relations, the spin polarization and distribution of spin among the two bands, and the nanotube chirality. Nevertheless, using reasonable parameters our model shows that it is

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possible to obtain a much higher value for δR in the ferrmagnetic/nanotube device (up to 0.25) than is predicted by the diffusive model.[3,4] In addition, large oscillations in δR are predicted as a function of energy. These might be observable by measuring the nanotube magnetoresistance as a function of bias on a capacitively coupled gate, thus forming a new kind of nanotube spin transistor.

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