Temperature-mediated switching of magnetoresistance in Co-contacted multiwall carbon nanotubes

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We present temperature-dependent measurements of the magnetoresistance in ferromagnetically contacted multiwall nanotubes. At low temperature, the resistance increases sharply near zero-field due to misalignment of the contact magnetizations. As temperature increases, the resistance peak transforms into a resistance dip, with a peak-to-valley ratio of similar magnitude, but opposite sign. The resistance switch has a distinct temperature dependence compared with the background magnetoresistance, suggesting that the two have different origins. We propose that a ferromagnetic transition near the contact interfaces reverses the polarity of the injected spin, and changes the sign of the resistance switch. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1597965]

Spin-polarized electron transport through carbon nanotubes is of great interest for the development of future spin-electronic devices.[1] Electrons travel up to 10 μm or more through nanotubes without losing momentum or spin information.[2] This makes large spin-mediated resistance changes possible. Tsukagoshi *et al.*[3] showed that the resistance of a cobalt contacted multiwall nanotube (MWNT) is dependent on the relative orientation of the magnetization of the ferromagnetic contacts, providing the evidence for spin polarized transport in carbon nanotubes. The resistance is low at high field when the magnetizations are parallel (R_p) and high around zero field when the contact magnetizations are antiparallel (R_a), resulting in a resistance peak near $B=0$. This result has since been confirmed in measurements of both single[1] and multiwall nanotubes.[4,5] Recently it has been shown that for certain ferromagnetically contacted MWNTs, R_p can actually be higher than R_a, resulting in the appearance of a resistance dip, rather than a resistance peak.[5] The conditions necessary for this surprising result are not yet understood.

Here we report on an intriguing temperature dependence of the resistance in ferromagnetically contacted nanotubes that helps shed light on the inverse switching, and spin injection into carbon nanotubes in general. We observe that the resistance switch can change sign from positive to negative as a function of increasing temperature. Stray field effects cannot account for this unusual temperature dependence, so that the results provide strong supporting evidence for spin polarized electron transport in carbon nanotubes. We propose that the sign change is due to the existence of a nonferromagnetic layer at the contact/nanotube interface whose thickness increases with increasing temperature, and that controls the polarity of the injected spin.

Details of our device fabrication have been previously reported.[1,3] Contacts to 20 nm diameter MWNTs are defined using electron beam lithography with a contact separation of approximately 200 nm. The contact material is a 70-nm-thick cobalt layer deposited using thermal evaporation. Two-terminal differential resistance measurements are made for temperatures between 1.5 and 175 K as a function of magnetic field directed perpendicular to the MWNT axis and parallel to the plane of the contacts. This configuration minimizes stray field effects.

The differential resistance is shown in Figs. 1(a) and 1(b) for two Co-contacted MWNTs, each at three different temperatures. The two devices were fabricated under similar conditions; however, the zero field resistance of the device in

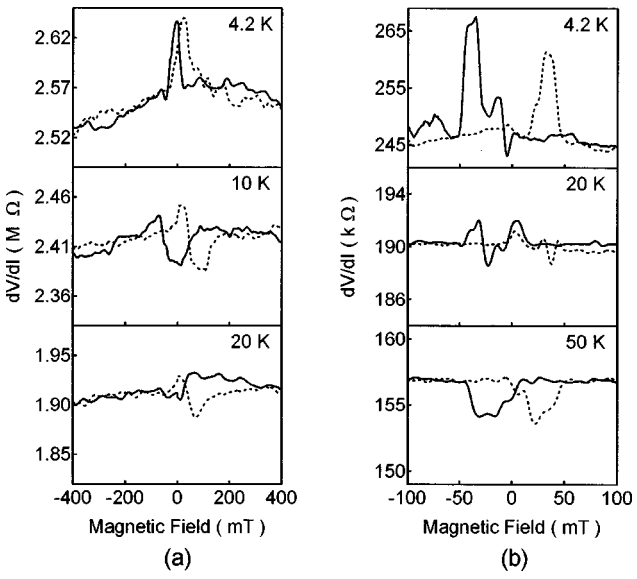

FIG. 1. Differential resistance as a function of magnetic field for (a) high resistance and (b) low resistance cobalt contacted MWNTs. The dashed (solid) line shows the forward (reverse) sweep direction. Magnetic field is directed perpendicular to the MWNT axis and parallel to the plane of the contacts.

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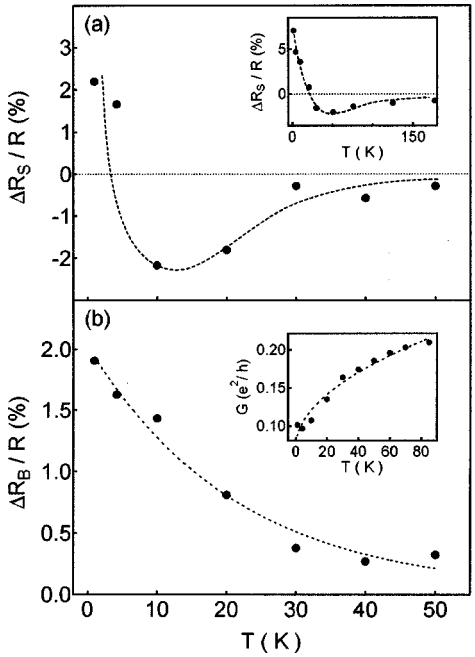

FIG. 2. (a) The resistance ratio $\Delta R_s/R=2(R_a-R_p)/(R_a+R_p)$ as a function of temperature for the high resistance device. Inset: $\Delta R_s/R$ as a function of temperature for the low resistance device. (b) The magnitude of the background resistance $\Delta R_B/R=2[R_{(B=100\ \mathrm{mT})}-R_{(B=500\ \mathrm{mT})}]/[R_{(B=100\ \mathrm{mT})}+R_{(B=500\ \mathrm{mT})}]$ as a function of temperature for the high resistance device. For each temperature, $\Delta R_B/R$ is averaged over the two sweep directions. Inset: The differential conductance as a function of temperature measured at 100 mT for the high resistance device.

Fig. 1(a) is approximately one order of magnitude higher than the resistance of the device in Fig. 1(b). This sample-to-sample variation is typical for nanotube devices, and suggests that tunnel barrier contacts are formed, in which small local variations in the interfacial barrier determine the total contact resistance.[6] At low temperatures, we observe a hysteretic resistance peak as the magnetic field sweeps from $+B$ to $-B$ and back to $+B$. This can be understood using the spin injection picture that has been described for magnetic tunnel junctions (MTJs).[3,7] Near $B=0$, the magnetization of the ferromagnetic contacts becomes misaligned. This results in a decrease in the overlap of the spin populations between the two contacts, causing the resistance to increase. As the field increases, the contact magnetizations realign, lowering the resistance back to the high field value. The result is a resistance peak at zero magnetic field. The hysteresis in the resistance peak reflects the hysteresis in the contact magnetizations.

In Fig. 2(a) and the inset to Fig. 2(a), the resistance ratio $\Delta R_s/R=2(R_a-R_p)/(R_a+R_p)$ is plotted as a function of temperature for the high- and the low-resistance device, respectively. The maximum $\Delta R_s/R$ is found to be 2.5% for the high-resistance device and 9% for the low-resistance device. As the temperature increases, the resistance peak progressively changes over into a resistance dip, after which the magnitude of the resistance dip gradually decreases to zero. The changeover from a peak to a dip occurs at approximately 10 K for the high-resistance device and at approximately 30 K for the low-resistance device, while the resistance switching survives up to 175 K for the low-resistance device and 50 K for the high-resistance device.

We first discuss the possible influence of stray magnetic field on the nanotube resistance. As the magnetizations of the ferromagnetic contacts change from the parallel to the antiparallel state, the magnetic field in the vicinity of the nanotube decreases, due to the partial cancellation of the stray field. The reduction in stray field can produce a peak in the measured resistance, since the MWNT has a background negative magnetoresistance[8] [seen in Fig. 1(a)]. According to previously reported magnetostatic calculations, the change in stray field is not large enough to produce the relatively large resistance switches observed in our ferromagnetically contacted nanotube devices.[9] The present data provides additional compelling evidence that stray field effects can be neglected. In Fig. 2 we compare the temperature dependences in the high-resistance device for (a) the resistance switching $(\Delta R_s/R)$ and (b) the background resistance change $(\Delta R_B/R)$. If stray field effects are the cause of the resistance switching, we expect that the temperature dependencies of $\Delta R_s/R$ and $\Delta R_B/R$ should be identical. A fit to the data shows that $\Delta R_B/R$ decreases exponentially with increasing temperature. The magnitude of $\Delta R_s/R$, on the other hand, is approximately constant at low temperature, and then drops off rapidly near 30 K (a sharp drop-off in $\Delta R_s/R$ at a critical temperature was also reported in Ref. 4). The sign of $\Delta R_s/R$ meanwhile changes from positive to negative. The two distinctly different temperature dependences strongly suggest that stray field effects are not responsible for the resistance switching.

Within the spin injection picture, a resistance dip around zero field implies that electron transmission in the parallel spin configuration is lower than in the antiparallel configuration.[5] Recent theory has shown that this counterintuitive situation can be brought about by establishing the appropriate contact interface conditions.[10] In cobalt, negatively spin polarized *d* electrons and positively spin-polarized *s* electrons coexist at the Fermi energy. The relative coupling strength of the two electron populations across the ferromagnetic/nonferromagnetic interface determines the sign of the injected spin polarization. Experimentally, a thin Ru interface layer has been shown to reverse the polarization of spin injected from a cobalt contact leading to an inverse resistance switch in MTJs,[11] while MTJs made with $SrTiO_3$ or Ta_2O_5 tunnel barriers show inverse resistance switching at low bias.[12]

In our samples, the contact interface conditions transform to cause the resistance ratio to change sign as a function of increasing temperature. We propose that this is due to a ferromagnetic transition of a mixed composition layer at the nanotube-contact interface. Since cobalt oxidizes readily at room temperature, the interface between the cobalt contact and the nanotube consists of a mixture of cobalt–oxide and cobalt. Although on its own cobalt–oxide is antiferromagnetic, cobalt/cobalt–oxide mixtures can be ferromagnetic at low enough temperature. Magnetization measurements show that the spins in cobalt–oxide align with the ferromagnetic cobalt constituent, producing a large saturation magnetization.[13] The saturation magnetization disappears at a transition temperature that decreases with increasing percentage of cobalt oxide. In our samples, the percentage of cobalt oxide is highest at the nanotube interface, and de-

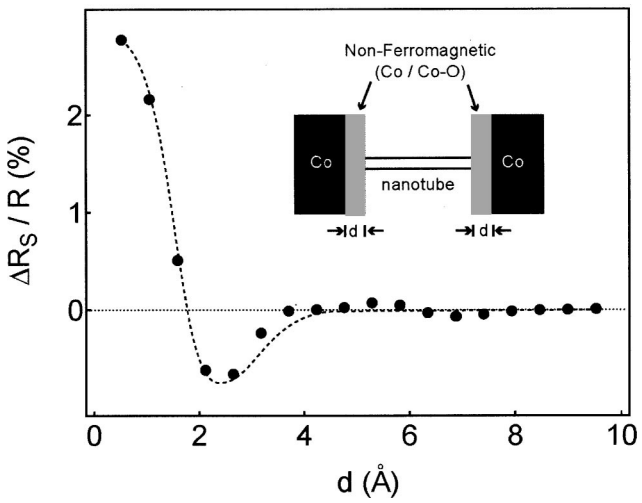

FIG. 3. The resistance ratio $\Delta R_s/R=2(T_p-T_a)/(T_a+T_p)$ calculated using a one-dimensional Schrödinger equation for a ferromagnetically contacted armchair nanotube, as a function of the width of non-ferromagnetic interface layers. In the cobalt contacts, the energy splitting between the spin-up and spin-down electrons $\Delta E=1.45$ eV, the Fermi energy $E_F=2.2$ eV and the effective mass $m^*=1m_e$ (see Ref. 16). Each nonferromagnetic region is divided into $n=d/d_0$ small sections with $d_0=1$ a.u.$=0.529$ Å. ΔE is taken to be constant in each small section but to decrease to zero in such a way that k changes linearly across the width of the nonferromagnetic regions, while E_F and m^* are kept constant as in the cobalt contacts. For the nanotube, the length $L=200$ nm, $k=0.8509$/Å and $m^*=1m_e$ are taken.

creases with increasing distance into the cobalt contact. As temperature increases then, a non-ferromagnetic layer forms within the contact/nanotube interface, and the thickness of this layer increases with increasing temperature (see inset to Fig. 3).

We have performed a model calculation to show how the presence of a nonferromagnetic layer within the contact/nanotube interfaces can change the relative transmission of the spin-up and spin-down electrons, and thus the sign of the resistance ratio. (A similar model has previously been applied to MTJs.)[14,15] The electron transmission through the nanotube device for the parallel (T_p) and antiparallel (T_a) configurations is calculated by solving the one-dimensional Schrödinger equation, taking into account the difference in the conduction band minimum ΔE for the spin-up and spin-down electrons in the ferromagnetic regions,[16] and assuming no spin scattering in the nanotube. The resistance ratio is then approximated by $\Delta R_s/R=2(T_p-T_a)/(T_a+T_p)$. The results of this calculation are plotted in Fig. 3 for a 200 nm long armchair nanotube with two ferromagnetic cobalt contacts, separated by two nonferromagnetic regions of variable width. As the width of the nonferromagnetic regions increase, the transmission coefficients T_a and T_p oscillate, each out of phase from one another, causing $\Delta R_s/R$ to change from positive to negative. For larger nonferromagnetic layer thickness, the phase shift between T_a and T_p decreases, and $\Delta R_s/R$ gradually goes to zero. As shown in Fig. 3, the calculation is in good qualitative agreement with our data. To quantitatively determine the resistance ratio—including the influence of the carbon nanotube electron state distribution on the sign of the injected spin—a detailed theory of the type described in Ref. 10 for ferromagnetic/nanotube junctions is needed. Nevertheless, our simple calculation demonstrates that an increase in the thickness of nonferromagnetic layers at the contacts as a function of increasing temperature could in fact produce the observed transition.

In summary, we have studied the temperature dependence of spin injection from ferromagnetic contacts into carbon nanotubes. The resistance switching is observed to change sign as a function of increasing temperature, indicating the existence of a nonferromagnetic interfacial layer that modifies the polarization of the injected spin. This implies that to observe larger and more reproducible spin mediated effects in carbon nanotube devices, improvements in the purity of the ferromagnetic interface layer are required.

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[1] B. Alphenaar, S. Chakraborty, and K. Tsukagoshi, *Electron transport in quantum dots* (Kluwer Academic, New York, 2003).
[2] C. T. White and T. N. Todorov, Nature (London) **393**, 240 (1998); S. J. Tans, M. H. Devoret, H. Dai, A. Thess, R. E. Smalley, L. J. Geerligs, and C. Dekker, *ibid.* **386**, 474 (1997); A. Batchtold, M. S. Fuhrer, S. Plyasunov, M. Foreo, E. H. Anderson, A. Zettl, and P. L. McEuen, Phys. Rev. Lett. **84**, 6082 (2000).
[3] K. Tsukagoshi, B. W. Alphenaar, and H. Ago, Nature (London) **401**, 572 (1999).
[4] D. Oragassa, G. J. Mankey, and H. Fujiwara, Nanotechnology **12**, 281 (2001).
[5] B. Zhao, I. Monch, T. Muhl, H. Vinzelberg, and C. M. Schneider, J. Appl. Phys. **91**, 7026 (2002); Appl. Phys. Lett. **80**, 3144 (2002).
[6] E. L. Wolf and D. L. Lossee, Phys. Rev. B **2**, 3660 (1970); A. Kanda, Y. Ootuka, K. Tsukagoshi, and Y. Aoyagi, Appl. Phys. Lett. **79**, 1354 (2001).
[7] G. A. Prinz, Science **282**, 1660 (1998).
[8] C. Schonenberger, A. Bachtold, C. Strunk, J.-P. Salvetat, and L. Forro, Appl. Phys. A: Mater. Sci. Process. **69**, 283 (1999).
[9] B. W. Alphenaar, K. Tsukagoshi, and M. Wagner, J. Appl. Phys. **89**, 6863 (2001).
[10] E. Yu. Tsymbal and D. G. Pettifor, J. Phys.: Condens. Matter **9**, L411 (1997); I. I. Oleink, E. Yu. Tsymbal, and D. G. Pettifor, Phys. Rev. B **62**, 3952 (2000).
[11] P. LeClair, B. Hoex, H. Wieldraaijer, J. T. Kohlepp, H. J. M. Swagten, and W. J. M. de Jonge, Phys. Rev. B **64**, 100406 (2001).
[12] J. M. De Teresa, A. Barthelemy, A. Fert, J. P. Contour, R. Lyonnet, F. Montaigne, P. Seneor, and A. Vaures, Phys. Rev. Lett. **82**, 4288 (1999); M. Shrama, S. X. Wang, and J. H. Nickel, *ibid.* **82**, 616 (1999).
[13] D. V. Dimitrov, G. C. Hadjipanayis, V. Papefthymiou, and A. Simopoulos, IEEE Trans. Magn. **33**, 4363 (1997).
[14] J. S. Moodera, J. Nowak, L. R. Kinder, and P. M. Tedrow, Phys. Rev. Lett. **83**, 3029 (1999).
[15] J. C. Slonczewski, Phys. Rev. B **39**, 6995 (1989).
[16] M. B. Stearns, J. Magn. Magn. Mater. **5**, 167 (1977).