

# Electrical spin injection in multiwall carbon nanotubes with transparent ferromagnetic contacts

S. Sahoo, T. Kontos,<sup>a)</sup> and C. Schönenberger

*Institut für Physik, Universität Basel, Klingelbergstrasse 82, CH-4056 Basel, Switzerland*

C. Sürgers

*Physikalisches Institut and DFG Center for Functional Nanostructures, University of Karlsruhe, D-76128 Karlsruhe, Germany*

(Received 30 August 2004; accepted 21 January 2005; published online 9 March 2005)

We report on electrical spin injection measurements on multiwall carbon nanotubes (MWNTs). We use a ferromagnetic alloy  $\text{Pd}_{1-x}\text{Ni}_x$  with  $x \approx 0.7$  which allows us to obtain devices with resistances as low as 5.6 k $\Omega$  at 300 K. The yield of device resistances below 100 k $\Omega$ , at 300 K, is around 50%. We measure at 2 K a hysteretic magneto-resistance due to the magnetization reversal of the ferromagnetic leads. The relative difference between the resistance in the antiparallel (AP) orientation and the parallel ( $P$ ) orientation is about 2%. © 2005 American Institute of Physics. [DOI: 10.1063/1.1882761]

How the spin degree of freedom can propagate and be manipulated in low-dimensional devices is a question of both fundamental and technical interest. On the one hand, proposals for a spin field-effect transistor (SpinFET),<sup>1</sup> which one can consider as a generic spintronic scheme, rely on electrical spin injection in one-dimensional channels. On the other hand, spin transport is expected to provide new information on the peculiar nature of an electronic fluid, as electron-electron interactions are enhanced when one reduces the dimensionality. Within the framework of the Luttinger liquid model, for example, Balents and Egger showed theoretically that spin-charge separation modifies qualitatively spin transport in quantum wires.<sup>2</sup>

Carbon nanotubes (NTs) can be considered as one-dimensional (1D) or zero-dimensional (0D) conductors, with important Coulomb interaction.<sup>3,4</sup> Spin transport is thus a powerful tool for the study of their intrinsic properties. Interestingly, in view of the conventional Elliot mechanism<sup>5</sup> for spin relaxation in metals, one expects a relatively long spin relaxation length (several  $\mu\text{m}$ ), because of the expected low spin-orbit coupling. This makes carbon nanotubes potentially attractive for device applications.

The main problem encountered in previous studies of electrical spin injection in NTs was to find ferromagnetic metals which can contact reliably the NTs, with a low ohmic device resistance.<sup>6-8</sup> A low device resistance is not *a priori* needed in a macroscopic spin valve, where the conductance is controlled by the relative orientation of the magnetization of two ferromagnetic electrodes around an insulating barrier. However, transparent ferromagnetic contacts on NTs are essential for the study of spin dependent transport at low temperatures, to avoid quenching transport because of charging effects.

In this letter, we present a scheme for contacting MWNTs with a ferromagnetic alloy,  $\text{Pd}_{1-x}\text{Ni}_x$ , with  $x \approx 0.7$ . Shape anisotropy is used for controlling the coercive field of the ferromagnetic contacts. This scheme allows us to achieve contact resistances of, on average, 30 k $\Omega$  at room temperature. The minimum devices resistance measured so far is

5.6 k $\Omega$ . The yield of devices with resistances below 100 k $\Omega$ , at 300 K, is around 50%. In the linear conductance regime, we find that the resistance switches hysteretically when sweeping the magnetic field. The relative difference between the resistance in the antiparallel (AP) orientation and the parallel ( $P$ ) orientation is about 2%.

Giant paramagnetism is a well-known feature of Pd and few magnetic impurities added to its matrix can drive it into the ferromagnetic state.<sup>11</sup> Therefore, as Pd alone makes quasi-adiabatic contacts on NTs,<sup>9</sup> ferromagnetic Pd alloys are expected to keep the same contacting properties as Pd, provided the concentration of magnetic impurities is low enough. However, as the spin signal is proportional to  $P^2$ ,  $P$  being the spin polarization of the alloy,<sup>10</sup> the concentration should not be too small to ensure that the current which is driven in the MWNTs is enough spin polarized. As we will see below, the contacting properties of  $\text{Pd}_{1-x}\text{Ni}_x$  on NTs remain very similar to pure Pd even in the case of high Ni concentration. Therefore, we chose to use the alloy in the concentrated limit ( $x \approx 0.7$ ) to ensure high enough spin polarization of the source-drain current.

The MWNTs used in this work are grown by arc discharge and stored as a suspension in chloroform. We first pre-pattern Au bonding pads and alignment marks on a thermally oxidized highly doped Si (resistivity of  $\approx 5 \text{ m}\Omega \cdot \text{cm}$  at 300 K), used as back-gate ( $\text{SiO}_2$  thickness  $\approx 400 \text{ nm}$ ). We spread the MWNTs on this substrate and localize them under a SEM (Philips FEG XL30 or LEO Supra 35). Using conventional e-beam lithography techniques, we write and develop the structure shown after lift-off in Fig. 1. In a vacuum system with a base pressure of about  $5.10^{-8}$  mbar, we first deposit a layer of  $\text{Pd}_{1-x}\text{Ni}_x$  with  $x \approx 0.7$  of 600 Å, at a pressure of about  $10^{-7}$  mbar. We use angle evaporation to obtain isolated ferromagnetic electrodes. We finally deposit 400 Å of Pd to connect the device to the pre-patterned Au bonding pads (not shown in Fig. 1). This Pd-PdNi bilayer is also evaporated on a bare substrate placed nearby in order to characterize the alloy by SQUID and vibrating sample magnetometry and Rutherford backscattering spectrometry (RBS). For all the samples for which we could study spin injection (six samples), the spacing of the ferromagnetic pads

<sup>a)</sup>Electronic mail: takis.kontos@unibas.ch

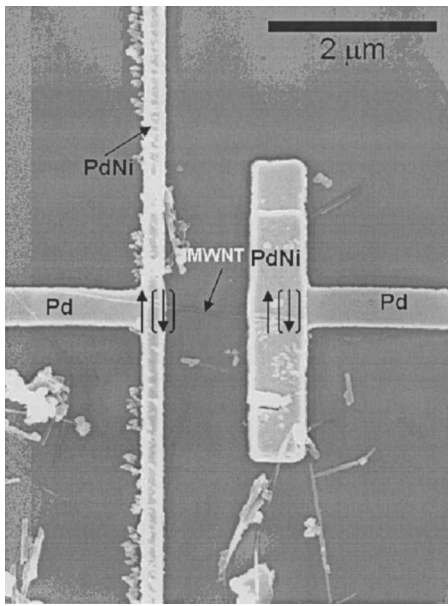


FIG. 1. A SEM picture of a device. The  $\text{Pd}_{0.3}\text{Ni}_{0.7}$  electrodes have different shapes,  $14\ \mu\text{m} \times 0.1\ \mu\text{m}$  and  $3\ \mu\text{m} \times 0.5\ \mu\text{m}$  for the left and the right electrode, respectively. They are spaced by  $1\ \mu\text{m}$ . The black arrows indicate the direction of the magnetization in the AP or the  $P$  orientation.

was either  $1\ \mu\text{m}$  or  $500\ \text{nm}$ . As shown in Fig. 1, the ferromagnetic electrodes have different shapes. This is to achieve different coercive fields for the two electrodes, by shape anisotropy, in order to produce a spin valve. Typical dimensions are  $14\ \mu\text{m} \times 0.1\ \mu\text{m}$  and  $3\ \mu\text{m} \times 0.5\ \mu\text{m}$  for the left and the right electrode, respectively.

In Figs. 2(a) and 2(b), we show the magnetic characterization of a thin film of  $600\ \text{\AA}$  of  $\text{Pd}_{1-x}\text{Ni}_x$  with  $x \approx 0.7$  under

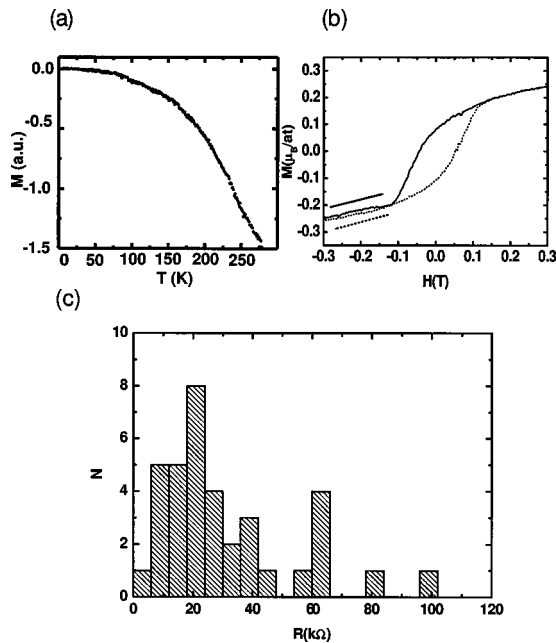


FIG. 2. (a) Temperature dependence of the magnetization of a thin film of  $\text{Pd}_{0.3}\text{Ni}_{0.7}$  of  $600\ \text{\AA}$  coated by  $500\ \text{\AA}$  of Pd, obtained while evaporating the contacts on the NT. The Curie temperature of the alloy is around  $270\ \text{K}$ . (b) Magnetic-field dependence of the magnetization of the  $\text{Pd}_{0.3}\text{Ni}_{0.7}$  film at  $T = 2.7\ \text{K}$ . As expected, a hysteresis is observed. The saturation magnetization is about  $0.25\ \mu_B$ . (c) Histogram of the contacting properties of PdNi on MWNTs at  $300\ \text{K}$ . The mean resistance is  $30\ \text{k}\Omega$  and the most probable one is  $20\ \text{k}\Omega$ .

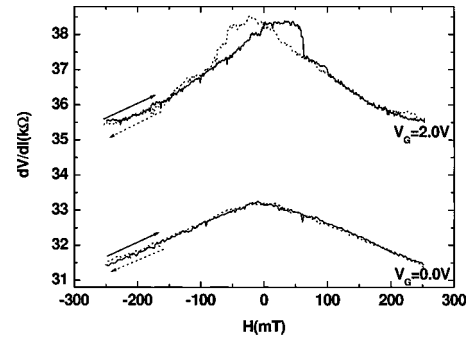


FIG. 3. Magnetic-field dependence of the linear resistance at  $1.85\ \text{K}$  as a function of an in-plane magnetic-field parallel to the axis of the ferromagnetic electrodes, for gate voltages  $V_G = 2.0\ \text{V}$  and  $V_G = 0.0\ \text{V}$ . A hysteretic behavior characteristic of a spin valve is observed for  $V_G = 2.0\ \text{V}$ .

$400\ \text{\AA}$  of Pd. The magnetic-field dependence of the magnetization displays a hysteresis loop with a coercive field of  $50\ \text{mT}$  (the field is parallel to the layer). The magnetization saturates at around  $0.25\ \mu_B$  per total number of atoms and decreases rapidly above  $270\ \text{K}$ . Note that, although this is enough to study spin transport below  $100\ \text{K}$ , the Curie temperature and the saturation magnetization are 50% smaller than the known bulk characteristics for this Ni concentration.<sup>11</sup> We think that this might be due to partial oxidation of the Ni during evaporation. Evaporating the alloy at a lower pressure could allow us to achieve room temperature ferromagnetic contacts. The Ni concentration in the Pd matrix is measured on the same thin film by RBS.

Figure 2(c) shows the histogram of the device resistances. On 100 NTs contacted so far, we could contact successfully 46 of them with a device resistance below  $100\ \text{k}\Omega$ . As shown on the histogram, the average device resistance of these 46 samples is around  $30\ \text{k}\Omega$  at  $300\ \text{K}$ . The lowest device resistance was found to be  $5.6\ \text{k}\Omega$  at  $300\ \text{K}$  and the most probable one is  $20\ \text{k}\Omega$ . All of these resistances are measured for a gate voltage  $V_G = 0.0\ \text{V}$  in the linear regime. At  $2\ \text{K}$ , the linear resistance remains typically below  $100\ \text{k}\Omega$  when sweeping the gate, which allows us to study spin transport.

The dependence of the linear resistance  $dV/dI$  of a device versus an applied magnetic-field  $H$  for two different gate voltages  $V_G = 2.00\ \text{V}$  and  $V_G = 0.00\ \text{V}$  is shown in Fig. 3. In order to take advantage of shape anisotropy, the field is kept parallel to the axis of the ferromagnetic pads. The overall behavior is a decrease of the resistance as one increases the magnetic field, as previously reported.<sup>12,13</sup> In addition, for  $V_G = 2.00\ \text{V}$ , the resistance displays a hysteretic behavior. Around  $0\ \text{mT}$ , it gradually increases further upon reversing the sign of the magnetic field and switches to a lower value around  $100\ \text{mT}$ . As expected for a spin valve, the two curves  $dV/dI(H)$  obtained when sweeping down or up match at high field and are roughly mirror-symmetric. Therefore, when reversing the sign of the magnetic field, the region between  $0$  and  $60\ \text{mT}$  corresponds to an antiparallel (AP) orientation of the magnetizations of the electrodes, whereas all the other regions of field correspond to the parallel ( $P$ ) orientation. We define the TMR as

$$\text{TMR} = 2 \frac{R_{\text{AP}} - R_P}{R_P + R_{\text{AP}}}$$

where  $R_{\text{AP}}$  is the resistance in the AP orientation at  $50\ \text{mT}$  and  $R_P$  is the resistance in the  $P$  orientation at the same field.

For  $V_G=2.0$  V, the TMR is positive, around 2.05%. Even though the exact spin polarization of the alloy is not known, one can roughly estimate it comparing the magnetization of the actual alloy with that of pure Ni. Taking the known value for the spin polarization  $P_{\text{Ni}}$  of Ni,<sup>14</sup> one obtains  $P_{\text{PdNi}} \approx \mu_{\text{PdNi}} P_{\text{Ni}} / \mu_{\text{Ni}} \approx 0.25 * 23 / 0.6 = 9.58\%$ . This spin polarization would yield a TMR of 1.85% for a tunnel junction within the simple Jullière's model.<sup>10</sup> Although this amplitude is in reasonable agreement with our measurements, this comparison probably underestimates spin-dependent and/or energy dependent scattering in the nanotube. For example, charging effects could be important. They are indeed observed in the spin independent part of the  $R$  versus  $V_G$  characteristic.

The resistance change of about 2% measured for  $V_G=2.0$  V could also be accounted for by a change of about 50 mT of local stray magnetic field arising from the ferromagnetic pads contacting the nanotube. For ruling out this spurious effect, one can define the sensitivity to external local magnetic fields of the nanotube-device as the slope of the  $R$  versus  $H$  curve when there is no magnetic switching. For  $V_G=2.0$  V, it is 0.037% /mT (14  $\Omega$ /mT) and for  $V_G=0.0$  V, it is 0.018% /mT (6  $\Omega$ /mT). Thus, a change in the stray fields of 50 mT would indeed change the resistance at  $V_G=2.0$  V of about 2% but would also change the resistance at  $V_G=0.0$  V by 1%. However, as shown in Fig. 3, there is a hysteresis lower than 0.1% in the  $R$  versus  $H$  curve for  $V_G=0.0$  V, whereas a hysteresis of 2% is present for  $V_G=2.0$  V. We can, therefore, rule out stray magnetic-field effects from the ferromagnetic pads, as they should be independent of the gate voltage. The electronic current flowing through the tube is spin polarized. Note also that Fig. 3 shows that the TMR is gate controlled. This gate dependence is presently not understood and will be studied in subsequent papers.

In conclusion, we have demonstrated reliable contacting and spin injection in MWNTs with transparent contacts. Using a  $\text{Pd}_{1-x}\text{Ni}_x$  alloy with  $x \approx 0.7$ , we can have device resistances as low as 5.6 k $\Omega$  at 300 K. At 2 K, we observe a TMR of about 2%. We think that this contacting scheme will allow extensive studies of spin effects in NTs in the 0D or the 1D regime and can be used in principle for device applications.

The authors acknowledge fruitful discussions with R. Al-lenspach, W. Belzig, and A. Cottet. The authors thank F. Lulu and M. Aprili for the RBS measurements. The authors thank Lazlo Forró for the MWNTs. This work is supported by the DIENOW RTN network, the NCCR on nanoscience and by the Swiss National Foundation.

<sup>1</sup>S. Datta and B. Das, Appl. Phys. Lett. **56**, 665 (1990).

<sup>2</sup>L. Balents and R. Egger, Phys. Rev. B **64**, 035310 (2001).

<sup>3</sup>S. J. Tans, R. M. Verschueren, and C. Dekker, Nature (London) **394**, 761 (1998).

<sup>4</sup>M. Bockrath, D. H. Cobden, J. Lu, A. G. Rinzler, R. E. Smalley, L. Balents, and P. L. McEuen, Nature (London) **397**, 598 (1999).

<sup>5</sup>I. Zutic, J. Fabian, and S. Das Sarma, Rev. Mod. Phys. **76**, 323 (2004).

<sup>6</sup>K. Tsukagoshi, B. W. Alphenaar, and H. Ago, Nature (London) **401**, 572 (1999).

<sup>7</sup>J. R. Kim, H. M. So, J. J. Kim, and J. Kim, Phys. Rev. B **66**, 233401 (2002).

<sup>8</sup>B. Zhao, I. Mönch, H. Vinzelberg, T. Mühl, and C. M. Schneider, Appl. Phys. Lett. **80**, 3144 (2002).

<sup>9</sup>A. Javey, J. Guo, Q. Wang, M. Lundstrom, and H. Dai, Nature (London) **424**, 654 (2003). We have developed independently the contacting with Pd and PdNi. See B. Babic, T. Kontos, and C. Schönenberger, Phys. Rev. B **70**, 235419 (2004).

<sup>10</sup>M. Jullière, Phys. Lett. **54A**, 225 (1975).

<sup>11</sup>J. Beille, Ph. D. thesis, Université Joseph Fourier, Grenoble, 1975.

<sup>12</sup>A. Bachtold, Christoph Strunk, Jean-Paul Salvetat, Jean-Marc Bonard, Laszl Forró, Thomas Nussbaumer, and Christian Schönenberger, Nature (London) **397**, 673 (1999).

<sup>13</sup>C. Schönenberger, A. Bachtold, C. Strunk, and J.-P. Salvetat, Appl. Phys. A: Mater. Sci. Process. **69**, 283 (1999).

<sup>14</sup>R. Meservey and P. M. Tedrow, Phys. Rep. **238**, 173 (1994).