Electrical Spin Injection in Multi-Wall carbon NanoTubes with transparent ferromagnetic contacts

S. Sahoo, T. Kontos and C. Schönenberger
Institut für Physik, Universität Basel,
Klingelbergstrasse 82,
CH-4056 Basel, Switzerland.

C. Sürgers
Physicalishes Institut and DFG Center for Functional Nanostructures,
University of Karlsruhe, D-76128 Karlsruhe, Germany
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Abstract

We report on electrical spin injection measurements on Multi-Wall carbon NanoTubes (MWNTs). We use a ferromagnetic alloy Pd$_{1-x}$Ni$_x$ with $x \approx 0.7$ which allows to obtain devices with resistances as low as 5.6 kΩ at 300 K. The yield of device resistances below 100 kΩ, 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%.

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How the spin degree of freedom propagates and can be manipulated in low dimensional
devices is a question of both fundamental and technical interest. On one hand, proposals for
a spin field-effect transistor (SpinFET) [1], which one can consider as a generic spintronic
scheme, rely on electrical spin injection in 1-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 [2].

Carbon nanotubes (NTs) can be considered as 1-dimensional or 0-dimensional conduc-
tors, with important Coulomb interaction [3, 4]. Spin transport is thus a powerful tool
for the study of their intrinsic properties. Interestingly, in view of the conventional Elliot
mechanism [5] for spin relaxation in metals, one expects a relatively long spin relaxation
length (several \( \mu m \)), 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 [6, 7, 8]. 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,
Pd\(_{1-x}\)Ni\(_x\), with \( x \approx 0.7 \). Shape anisotropy is used for controlling the coercive field of the
ferromagnetic contacts. This scheme allows 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 [11]. Therefore, as Pd alone makes
quasi-adiabatic contacts on NTs [9], 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 [10], 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
Pd$_{1-x}$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 $\approx 5 \, m\Omega.cm$ at 300 $K$), used as back-gate (SiO$_2$
thickness $\approx 400 \, nm$). We spread the MWNTs on this substrate and localize them under
a SEM (Philips FEG XL30 or LEO). Using conventional e-beam lithography techniques,
we write and develop the structure shown after lift-off on figure ???. In a vacuum system
with a base pressure of about $5.10^{-8} \, mbar$, we first deposit a layer of Pd$_{1-x}$Ni$_x$ with $x 
\approx 0.7$ of 600 $\AA$, at a pressure of about $10^{-7} \, mbar$. We use angle evaporation to obtain
isolated ferromagnetic electrodes. We finally deposit 400 $\AA$ of Pd to connect the device to
the pre-patterned Au bonding pads (not shown on figure ??). This Pd/PdNi bilayer is also
evaporated on a bare substrate placed nearby in order to characterize the alloy by SQUID
magnetometry and RBS (Rutherford Backscattering Spectrometry). For all the samples for
which we could study spin injection (6 samples), the spacing of the ferromagnetic pads was
either 1 $\mu m$ or 500 $nm$. As shown on figure ??, 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 $\mu m \times 0.1 \, \mu m$ and 3 $\mu m \times 0.5
\mu m$ for the left and the right electrode respectively.

On figure ??a and b, we show the magnetic characterization of a thin film of 600 $\AA$ of
Pd$_{1-x}$Ni$_x$ with $x \approx 0.7$ under 400 $\AA$ of Pd. The magnetic field dependence of the magneti-
zation displays a hysteresis loop with a coercive field of 50 $mT$ (the field is perpendicular to
the layer). The magnetization saturates at around 0.25 $\mu_B$ per total number of atoms and
decreases rapidly above 270 $K$. Note that, although this is enough to study spin transport
below 100 $K$, the Curie temperature and the saturation magnetization are 50% smaller than
the known bulk characteristics for this Ni concentration [11]. We think that this might be
due to partial oxidation of the Ni during evaporation. Evaporating the alloy at a lower pressure could allow to achieve room temperature ferromagnetic contacts. The Ni concentration in the Pd matrix is measured on the same thin film by RBS.

Figure ??c shows the histogram of the device resistances. On the hundred of NTs contacted so far, we could contact successfully 46 of them with a device resistance below 100 kΩ. As shown on the histogram, the average device resistance of these 46 samples is around 30 kΩ at 300 K. The lowest device resistance was found to be 5.6 kΩ at 300 K and the most probable one is 20 kΩ. All these resistances are measured for a gate voltage $V_G = 0.0$ V in the linear regime. At 2 K, the linear resistance remains typically below 100 kΩ when sweeping the gate, which allows 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 $V_G = 2.00$ V and $V_G = 0.00$ V is shown on figure ???. 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 [12, 13]. In addition, for $V_G = 2.00$ V, the resistance displays a hysteretic behavior. Around 0 mT, it gradually increases further upon reversing the sign of the magnetic field and switches to a lower value around 100 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 mT and 60 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

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

where $R_{AP}$ is the resistance in the AP orientation at 50 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 $P_{Ni}$ of Ni[14], one obtains $P_{PdNi} \approx \mu_{PdNi} P_{Ni} / \mu_{Ni} \approx 0.25 \times 23 / 0.6 = 9.58\%$. This spin polarization would yield a TMR of 1.85 % for a tunnel junction within the simple Jullière’s model [10]. 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 $R$ vs $V_G$ 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 $R$ vs $H$ 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 on figure ??, there is a hysteresis lower than 0.1 % in the $R$ vs $H$ curve for $V_G = 0.0\, V$, whereas a hysteresis of 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 figure ?? 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 Pd$_{1-x}$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.

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[1] S. Datta and B. Das, Appl. Phys. Lett. 56, 665 (1990).

[2] L. Balents and R. Egger, Phys. Rev. B 64, 035310 (2001).

[3] S.J. Tans, R.M. Verschueren and C. Dekker, Nature, 394, 761 (1998).

[4] M. Bockrath, D.H. Cobden, J. Lu, A.G. Rinzler, R.E. Smalley, L. Balents, P.L. McEuen, Nature, 397, 598 (1999).

[5] I. Zutic, J. Fabian and S. Das Sarma, Rev. Mod. Phys., 76, 323 (2004).

[6] K. Tsukagoshi, B.W. Alphenaar and H. Ago, Nature, 401, 572 (1999).

[7] J.R. Kim, H.M. So, J.J. Kim and J. Kim, Phys. Rev. B 66, 233401 (2002).

[8] B. Zhao, I. Mönch, H. Vinzelberg, T. Mühl, and C. M. Schneider, Appl. Phys. Lett. 80, 3144 (2002).

[9] A. Javey et al., Nature 424, 654 (2003). We have developed independently the contacting with Pd and PdNi. See B. Babic, T. Kontos and C. Schönberger, cond-mat/0406626 and B. Babic, Proc. of the XVII Winterschool, Kirchberg, Austria, (2003).

[10] M. Jullière, Phys. Lett. 54A, 225 (1975).

[11] J. Beille, Ph. D. thesis, Université Joseph Fourier, Grenoble, 1975.

[12] A. Bachtold, Christoph Strunk, Jean-Paul Salvetat, Jean-Marc Bonard, Laszló Forr, Thomas Nussbaumer and Christian Schönberger, Nature, 397, 673 (1999).

[13] C. Schönberger, A. Bachtold, C. Strunk, and J.-P. Salvetat, Appl. Phys. A, 69, 283, (1999).

[14] R. Meservey and P.M. Tedrow, Phys. Rep. B 238, 173 (1994).
FIG. ?? A SEM picture of a device. The Pd$_{0.3}$Ni$_{0.7}$ electrodes have different shapes, 14 \( \mu m \times 0.1 \mu m \) and 3 \( \mu m \times 0.5 \mu m \) for the left and the right electrode respectively. They are spaced by 1 \( \mu m \). The black arrows indicate the direction of the magnetization in the AP or the P orientation.

FIG.?? a. Temperature dependence of the magnetization of a thin film of Pd$_{0.3}$Ni$_{0.7}$ of 600 Å coated by 500Å of Pd, obtained while evaporating the contacts on the NT. The Curie temperature of the alloy is around 270 \( K \). b. Magnetic field dependence of the magnetization of the Pd$_{0.3}$Ni$_{0.7}$ film at \( T = 2.7 K \). 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 300K. The mean resistance is 30 \( k\Omega \) and the most probable one is 20 \( k\Omega \).

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