

## Spin-dependent quantum interference in single-wall carbon nanotubes with ferromagnetic contacts

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We report the experimental observation of spin-induced magnetoresistance in single-wall carbon nanotubes contacted with high-transparency ferromagnetic electrodes. In the linear regime the spin-induced magnetoresistance oscillates with gate voltage in quantitative agreement with calculations based on a Landauer-Büttiker model for independent electrons. Consistent with this interpretation, we find evidence for bias-induced oscillation in the spin-induced magnetoresistance signal on the scale of the level spacing in the nanotube. At higher bias, the spin-induced magnetoresistance disappears because of a sharp decrease in the effective spin polarization injected from the ferromagnetic electrodes.

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Carbon nanotubes (NTs) with ferromagnetic contacts are a very rich and promising system for spintronics.<sup>1</sup> Depending on the transparency of the contacts, electronic transport through NTs occurs in different regimes. For low transparency a NT behaves as a quantum dot and Coulomb blockade determines the transport properties,<sup>2</sup> whereas for high transparency transport is mainly determined by quantum interference.<sup>3</sup> This versatility gives experimental access to spin-dependent transport phenomena that could not be investigated until now.<sup>4</sup>

Particularly important is the possibility of turning transport electrostatically, by means of a gate voltage. The gate-voltage dependence of spin transport through NTs has been studied only recently, with different experiments resulting in different experimental observations.<sup>5-7</sup> For instance, a significant but *gate-independent* influence of the contact magnetization on the NT conductance has been observed in Ref. 5. By contrast, the experiments from Ref. 6 show that the magnetoresistance attributed to the spin degree of freedom is gate-voltage dependent, but no correlation between spin-induced magnetoresistance and the linear conductance was found. Finally, Ref. 7 describes a magnetoresistance whose gate-voltage dependence correlates with the gate-voltage dependence of the linear conductance. For this last experiment, a full quantitative theory based on the Landauer-Büttiker picture was shown to describe the spin- and gate-dependent magnetotransport data.<sup>8</sup> Nevertheless, diverse phenomenology observed in the different experiments calls for additional experiments, possibly addressing different transport regimes and measurements at finite bias, to provide an unambiguous validation of a Landauer-Büttiker description of spin transport through NTs. These experiments are also important to convincingly exclude possible experimental artifacts that could mimic the spin-induced magnetoresistance, such as the magneto-Coulomb effect.<sup>9</sup>

Here, we report experimental investigations of spin transport through individual single-wall (SW) nanotubes with highly transparent ferromagnetic contacts. In this regime, which is qualitatively different from the one previously investigated (i.e., a SWNT quantum dot<sup>7</sup>), the gate- and bias-voltage-dependent conductance exhibits a so-called Fabry-

Pérot interference pattern, originating from quantum interference of electronic waves.<sup>10</sup> At low bias, we observe a clear spin-induced magnetoresistance (SIMR) whose magnitude oscillates periodically with the gate voltage. We show that the experimental data in the linear regime compare well to predictions based on a Landauer-Büttiker model for independent electrons and, at finite energy, we find experimental evidence for the expected bias-induced oscillations of the SIMR signal. At finite energy, we also find that the magnitude of the SIMR signal rapidly decreases when the bias is increased on a scale comparable to the exchange energy in the ferromagnetic electrodes. This indicates that in actual devices the effect of a bias-dependent spin polarization in the contacts needs to be taken into account.

The devices used in our investigations consist of SWNTs (either individual or ropes) in between two PdNi contacts, prepared on a degenerately doped silicon substrate (acting as a gate), coated with a 500-nm-thick thermally grown oxide layer. The SWNTs are deposited by means of a chemical vapor deposition process.<sup>11,12</sup> The electrodes consist of narrow strips ( $\sim 100$  nm) of a palladium-nickel alloy electron beam evaporated at a base pressure of  $10^{-7}$  mbar from a mixture of palladium and nickel in a 60% (Pd) to 40% (Ni) weight ratio. The alloy is ferromagnetic above room temperature.<sup>13</sup> As the spin polarization of pure nickel is 45%, the spin polarization of our PdNi alloy is lower than 20%, and estimated to be  $\approx 10\%$ . Despite this rather low polarization, the use of PdNi is advantageous, since the presence of Pd enables the realization of highly transparent contacts to the NTs.<sup>14,15</sup> We have investigated a number of different samples, and the data that we discuss here were measured at 4.2 K on a sample that was sufficiently stable to perform investigations of transport as a function of gate voltage, bias, and magnetic field.

The dependence of the sample conductance on gate and bias voltage is shown in Fig. 1(a). The conductance oscillates in a regular way as a function of both voltages. The resulting pattern is similar to that of Fabry-Pérot interference, which is typically observed in ballistic SWNTs, in the presence of backscattering at the contacts.<sup>10</sup> Spin-dependent transport in this Fabry-Pérot regime has not been addressed by previous

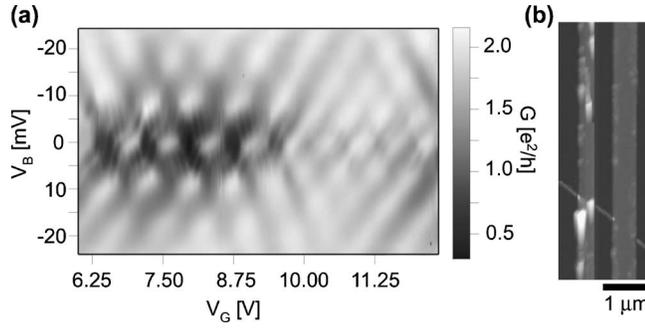


FIG. 1. (a) Gate and bias dependence of the conductance measured at 4.2 K, exhibiting a characteristic (Fabry-Pérot) pattern due to quantum interference of the electron waves (Ref. 10) (b) Atomic force microscope picture of the sample on which the data shown in this paper were measured. The obtained level spacing is approximately 6 mV, in reasonable agreement with the length of the nanotube between the contacts (350 nm).

investigations, which have only considered the case of Coulomb blocked SWNTs.<sup>5,7</sup>

Prior to investigating spin transport through NTs we have characterized the switching behavior of the ferromagnetic PdNi contacts by means of anisotropic magnetoresistance measurements. We found that the magnetization of PdNi lies approximately in the plane perpendicular to the strip, with a small component ( $\approx 10\text{--}15\%$  of the total magnetization) pointing along the strip. Sweeping the magnetic field in the plane perpendicular to the strip results in a magnetization reversal for  $|\mu_0 H|$  in between 100 and 300 mT, with the exact value depending on the width of the strip. At low field, a small, continuous change of the magnetization orientation also occurs due to the change of the in-plane component, since the complete alignment of the magnetization along an applied magnetic field requires a field of several tesla. For the SIMR measurements on SWNTs this behavior of the magnetization should result in hysteretic features at the magnetic field values for which the magnetization reversal occurs, superimposed onto smooth changes in the resistance due to the gradual change in magnetization orientation in the two PdNi leads.<sup>16</sup>

Figure 2(a) shows magnetoresistance curves for the SWNT sample measured for different values of the gate voltage, for an applied magnetic field increasing from  $-700$  to  $700$  mT (upsweep) and subsequently decreasing from  $700$  to  $-700$  mT (downsweep). The widths of the two PdNi contacts in this sample are 150 and 500 nm, resulting in magnetization reversal at magnetic field values of 250 and 125 mT, respectively. A hysteretic feature in the resistance is seen in the expected magnetic field range, corresponding to an antiparallel orientation of the magnetization in the PdNi electrodes. The feature is superimposed on a smoother background, as expected. The absolute change in resistance has a magnitude of a few tenths of a kilo-ohm, which is much larger than the total resistance of the PdNi strips. This implies that the effect cannot be accounted for in terms of a change in the resistance of part of the PdNi contacts, which is much lower. The change in magnetoresistance induced by the magnetization reversal is positive for most values of gate

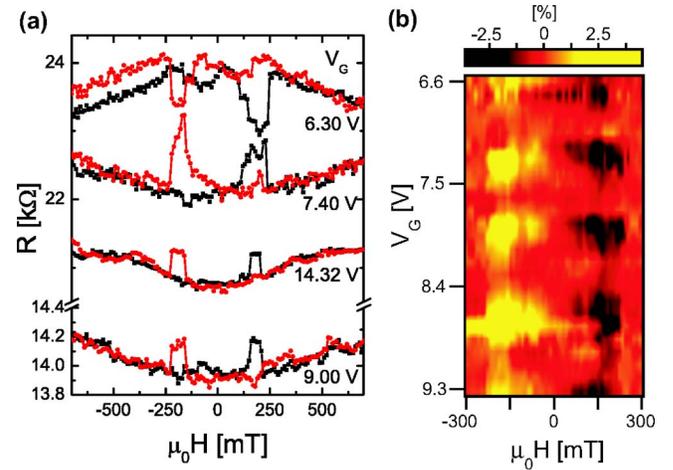


FIG. 2. (Color online) (a) Magnetoresistance traces measured at different gate voltages (black dots correspond to magnetic-field upsweeps; gray (red) dots to downsweeps). The reversal of the magnetization in the electrodes produces an increase in magnetoresistance; in a few cases a decrease is observed (e.g.,  $V_G = 6.30$  V). (b) Difference between the magnetoresistance signal measured in the up- and downsweeps, normalized to  $R_p = R(700 \text{ mT})$ , as a function of magnetic field and gate voltage. The periodicity with gate voltage is apparent.

voltage, i.e., the antiparallel orientation of the magnetization in the contacts results in an increase of the device resistance. In a few cases, however, a negative magnetoresistance has been observed<sup>7,8</sup> [see Fig. 2(a), top curve].

To investigate the SIMR in more detail, we have measured up- and downsweep linear magnetoresistance traces for  $V_G$  between 6.6 and 9.3 V, corresponding to approximately four periods of the gate-voltage-induced conductance oscillations. Figure 2(b) shows the difference between the resistance measured in the up- and down-magnetic-field sweeps as a function of  $V_G$  and  $\mu_0 H$ , normalized to the resistance at  $\mu_0 H = 700$  mT. It is apparent that the magnitude of the SIMR oscillates with gate voltage *in phase* with the resistance, which excludes the possibility that the observed magnetoresistance is due to a magneto-Coulomb effect.<sup>9</sup> The period of the oscillations also corresponds to that of the total conductance, thus demonstrating that the SIMR originates from quantum interference of electron waves backscattered at the contacts.

From the magnetoresistance measurements we also extract the conductance  $G_p = G(V_G, 700 \text{ mT})$  and the relative magnitude of the SIMR  $[R_{AP}(V_G) - R_p(V_G)]/R_p(V_G)$  (see Fig. 3). For  $R_{AP}(V_G)$  we take the value of the resistance measured at  $\mu_0 H = 175$  mT, i.e., in the middle of the hysteretic feature originating from the magnetization reversal. For  $R_p(V_G)$  we take the value measured at  $\mu_0 H = 700$  mT.

To interpret the linear magnetoresistance data, we adopt a Landauer-Büttiker picture. The spin-dependent, zero-temperature conductance of an individual SWNT can then be written as a function of the transmission probability for the majority spin (+) and minority spin (−) orientations of the left (*s*) and right (*r*) contacts as

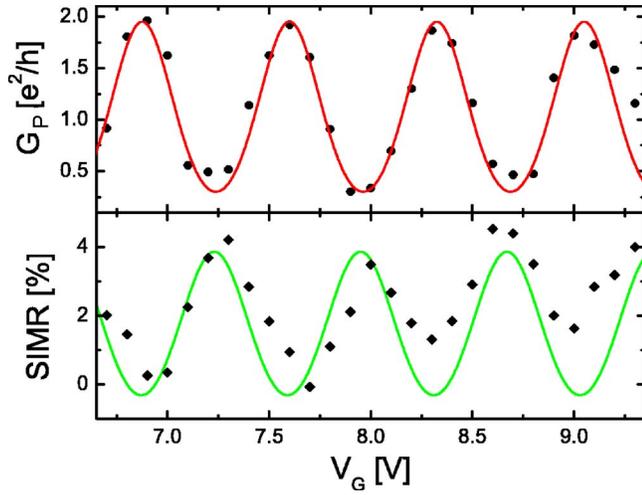


FIG. 3. (Color online) Comparison of theoretical predictions based on Eqs. (1) and (2) (lines) and experimental data (symbols) for the gate-voltage-dependent conductance (top) and amplitude of the spin-induced magnetoresistance (bottom).

$$G = \frac{2e^2}{h} \sum_{s,r \in \{+,-\}} T_{sr} \quad (1)$$

(the factor of 2 accounts for the doubly degenerate bands of SWNTs). Since in SWNTs spin-orbit interaction is negligibly small, we only have to consider transmission from  $(+)_s$  to  $(+)_r$  and  $(-)_s$  to  $(-)_r$  when the magnetization directions in the contacts are parallel to each other [i.e.,  $G_p = 2e^2/h(T_{++} + T_{--})$ ], and  $(+)_s \rightarrow (-)_r$  and  $(-)_s \rightarrow (+)_r$  spin [i.e.,  $G_{AP} = 2e^2/h(T_{+-} + T_{-+})$ ] when the magnetization vectors are antiparallel.

For ballistic propagation in the SWNT, the transmission probabilities read

$$T_{sr} = \frac{T_L^s T_R^r}{|1 - [(1 - T_L^s)(1 - T_R^r)]^{1/2} e^{2i\delta}|^2}, \quad (2)$$

where, following Refs. 7 and 8, we write the spin-dependent transmission probabilities  $T_{L(R)}^s$  at the left and right contacts as  $T_{L(R)}^s = T_{L(R)}(1 + sP)$ , with  $s = \pm 1$  and  $P$  the magnitude of the polarization in the contacts ( $P = 0.1$  as is appropriate for our PdNi electrodes). The quantity  $\delta$  is the phase acquired by one electron during its propagation from one contact to the other and back. It includes the spin-dependent phase acquired during the scattering process,<sup>8</sup> and the dynamical phase, which is linearly dependent on gate voltage and electron energy. We set the phase acquired in the reflections at the contacts to zero, to decrease the number of fitting parameters.

By convoluting Eq. (1) with the Fermi distribution, we evaluate the finite-temperature conductance and SIMR signal, using  $T_{L(R)}^s$  as the only free parameters. The solid lines in Fig. 3 correspond to the theoretical predictions obtained with  $T_L = 0.84$  and  $T_R = 0.26$ , i.e., the contacts exhibit only a small asymmetry. It is apparent that the model reproduces correctly the observed behavior of the linear conductance, the absolute

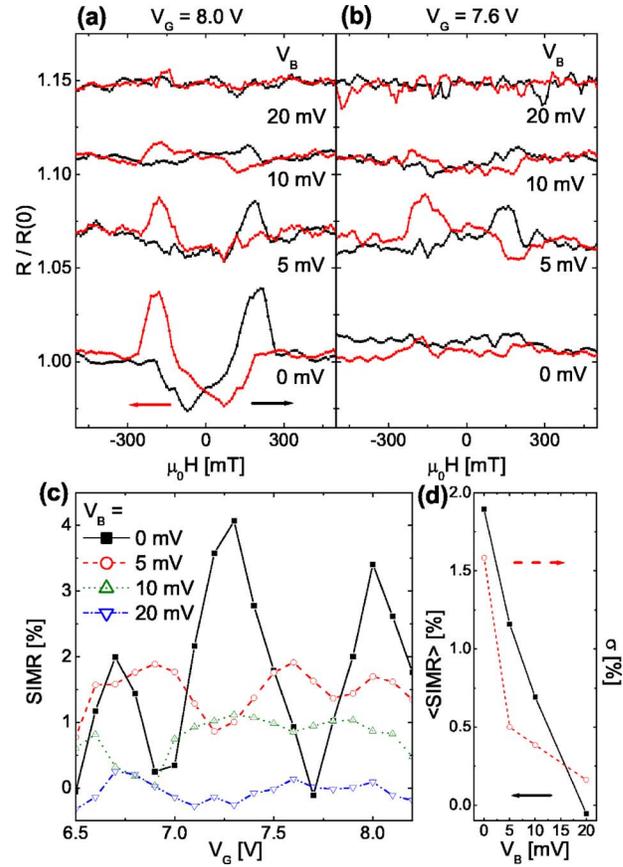


FIG. 4. (Color online) (a) and (b) Magnetoresistance traces for different values of the applied bias  $V_B$ , at  $V_G = 8.0$  and  $7.6$  V (curves offset for clarity). At  $V_G = 8.0$  V the SIMR signal at  $V_B = 0$  mV is larger than at  $V_B = 5$  mV; at  $V_G = 7.6$  V the situation is reversed. This indicates that the amplitude of the SIMR signal oscillates with bias. The full  $V_G$  dependence is shown in (c). (a)–(c) also clearly show that the SIMR signal at  $V_B = 20$  mV is fully suppressed. (d) Bias dependence of the gate-voltage-averaged SIMR and of its standard deviation (as a measure of the oscillating component).

magnitude of the SIMR signal ( $\sim 4\%$ ), and its periodicity for the entire measured gate-voltage range with only these two parameters. The small quantitative deviations seen in the detailed gate-voltage dependence of the SIMR signal can be explained (at least in part) by the misorientation of the magnetization in the two electrodes.

To understand to what extent a Landauer-Büttiker picture describes spin-dependent transport in SWNTs, we have also performed SIMR measurements at finite energy. The magnitude of the SIMR effect measured in the differential conductance should oscillate as a function of bias for independent electrons in a SWNT in the Fabry-Pérot regime, since the electron energy enters the phase  $\delta$  linearly, similarly to the gate voltage. Figures 4(a) and 4(b) show magnetoresistance traces measured at  $V_B = 0$  and 5 mV, for two values of the gate voltage. Whereas at  $V_G = 8.0$  V the SIMR signal at  $V_B = 0$  is larger than that at  $V_B = 5$  mV, at  $V_G = 7.6$  V the opposite is observed. This shows that oscillations as a function of bias on the scale of the level spacing in the nanotube (see Fig. 1) are indeed present, as further illustrated in Fig. 4(c) (curves at 0 and 5 mV), where the magnitude of the SIMR

signal is plotted versus  $V_G$ . This observation, in conjunction with the fact that also at finite bias the SIMR oscillates in phase with the gate-dependent differential resistance as expected from Eqs. (1) and (2), indicates that a Landauer-Büttiker picture provides a good first approximation to describe the energy dependence of spin-dependent transport in SWNTs.

Figure 4, however, also clearly shows that the magnitude of the SIMR signal rapidly decreases with increasing voltage. As a consequence, at  $V_B=20$  mV no SIMR is detected. Interestingly [see Fig. 4(d)], the rapid suppression with increasing bias occurs for both the  $V_G$ -dependent oscillations, originating from quantum interference, as well as for the gate-voltage-averaged signal, which corresponds to the “classical” SIMR signal. These observations deviate from the behavior expected on the basis of Eqs. (1) and (2) which predicts that the  $V_G$ -induced oscillations in the SIMR signal should preserve their amplitude at high bias. Observing that the classical (i.e., gate-averaged) SIMR signal disappears at high energy suggests that the suppression of SIMR with increasing bias does not originate from a breakdown of the Landauer-Büttiker picture, but rather from a decrease of the spin polarization in the electrodes, i.e., the polarization  $P$

decreases with increasing bias. This interpretation is suggested by the fact that the bias at which the SIMR signal disappears ( $\approx 20$  meV) is comparable to the exchange energy in the PdNi electrodes used in the experiments ( $E_{ex} \approx 30$  mV).<sup>13</sup> This is consistent with the known behavior of the magnetoresistance of tunnel junctions fabricated with many other weakly ferromagnetic alloys (e.g., Cu-Ni), which also show a rapid decrease of the effective spin polarization with increasing bias on the scale of the exchange energy.<sup>17</sup>

In conclusion, our systematic investigation of spin-dependent transport as a function of bias and gate voltage indicates that the observed phenomenology can be described in terms of a Landauer-Büttiker picture, provided that the bias dependence of the spin polarization injected from the ferromagnetic contacts is taken into account.

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