J Mater Sci (2011) 46:4614–4617
DOI 10.1007/s10853-011-5361-7

Spin transport in a thin graphite flake

J. Barzola-Quiquia · P. Esquinazi

Received: 4 November 2010 / Accepted: 31 January 2011 / Published online: 12 February 2011

Abstract We have studied the spin transport on a 30-nm thick and several micrometer long oriented graphite flake using a spin-valve configuration with four ferromagnetic Co electrodes of different widths and several μm separation. A 5-nm thin Pt layer has been introduced in between the ferromagnetic Co injector/detector and the graphite surface. In spite of the conductivity mismatch problem, efficient electrical spin injection and detection in graphite has been achieved. The magnetoresistance in the local and half-local electrodes shows clear maxima with symmetry around zero field. The spin transport can be detected up to 150 K.

Introduction

Due to the large electron mobility and mean free path of bulk graphite [1], thin crystalline graphite samples are expected to have some advantages for certain devices in comparison with single graphene layers fixed on dielectric substrates. Recent studies, for example, show that the graphene layers inside graphite have a smaller carrier density and a much larger mobility even at room temperature than single graphene layers [2]. That large spin diffusion can be expected in graphite should not actually be a surprise. Already doped Si is a material with relatively low carrier concentration and large spin diffusion length and efficiency [3, 4]. Nevertheless, the efficiency of spin injection between a ferromagnetic electrode and a graphite/graphene surface is a key point that needs to be studied and clarified. For example, theoretical calculations [5] indicate that the interfaces between graphite and (111) fcc or (0001) hcp Ni or Co surfaces should behave as perfect spin filtering. However, the formation of chemical bonds between graphite and the d-shell of those elements may diminish the spin injection. The authors of that theoretical work [5] proposed that the use of some Cu monolayers in between may preserve the spin and an ideal spin injection might be possible. In this study, we have tested the spin transport on a 30-nm thick and several micrometer long oriented graphite flake using a spin-valve configuration with four ferromagnetic (FM) Co electrodes of different widths and several μm large separation. In contrast to previous studies[6, 7], a 5-nm thin Pt layer has been deposited in between the ferromagnetic Co injector/detector layer and the graphite surface. In spite of the known conductivity mismatch problem, we show in this study that efficient electrical spin injection and detection in graphite with the described configuration is possible. The magnetoresistance in the local and half-local electrodes shows clear hysteresis with the common symmetry around zero field and with resistance maxima. The spin transport can be detected up to 150 K, overwhelming in temperature previous spin valve devices on mesoscopic graphite basis [6, 7].

Experimental details and sample characteristics

The samples were prepared by rubbing small flakes of highly ordered pyrolytic graphite (Advanced Ceramics) (grade ZYA, of rocking curve 0.4°) on a commercial 5 × 5 mm^2 silicon (100) substrate covered with a 150-nm SiN_x film; for the preparation details see [8]. For the measurements, we selected a graphite flake taking into account the

J. Barzola-Quiquia (✉) · P. Esquinazi
Division of Superconductivity and Magnetism, Institute for Experimental Physics II, University of Leipzig, Linnestr. 5, 04103 Leipzig, Germany
e-mail: j.barzola@physik.uni-leipzig.de

shape needed for the experiment, its thickness, and the Raman spectrum. After choosing the sample, we prepared the corresponding Au/Pt electrodes outside the sample area using conventional electron beam lithography. After that the Co/Pt FM electrodes (Pt directly on contact to the graphite surface) were prepared also by electron beam lithography in the conventional four points configuration. This process is done after the Au/Pt electrodes preparation in order to avoid possible oxidation of the Co film by the backing and developing process involved during the preparation of the lithography.

Resistance measurements were done in a commercial cryostat in the temperature range from 4 to 300 K using an AC Bridge (Linear Research LR-700). The magnetic field was parallel to the graphene planes of the graphite sample and along the long axes of the FM electrodes; it was measured using a Hall sensor fixed at the sample holder. In order to check the high-quality of the graphite flake, Micro-Raman spectrum was obtained at room temperature and ambient pressure with a Dilor XY 800 spectrometer at 514.53 nm wavelength (Green) and a 2 μm spot diameter. The incident power was kept at 1.5 mW to avoid possible sample damage or laser induced heating effects.

Figure 1 shows an electron microscope picture of the measured sample with the corresponding Co/Pt electrodes. In the experiments, we used four electrodes with two different widths in order to have different coercive fields. Figure 2 shows the Raman results. In graphite, three Raman peaks are of interest. Namely the G-peak around 1582 cm^{-1} due to the Raman active in-plane optical phonon, the D-peak around 1350 cm^{-1} that is very sensitive to

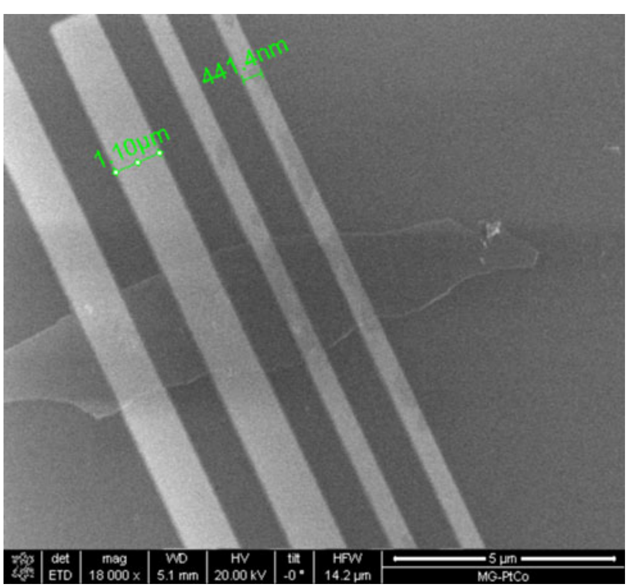

Fig. 1 Scanning electron microscope image of the investigated graphite flake with the four Co/Pt electrodes at the top. The length scale at the corner is 5 μm

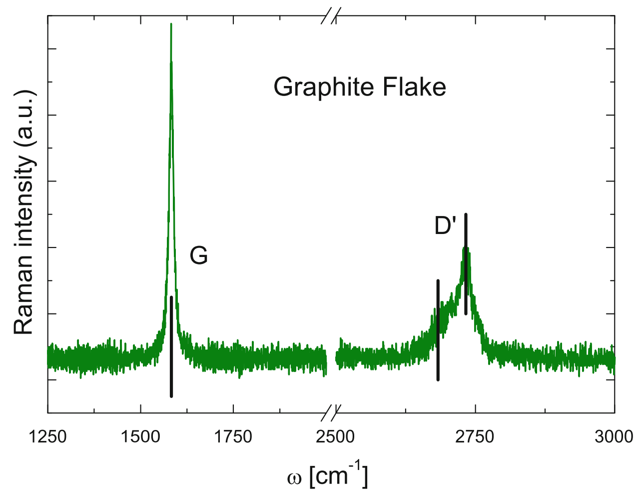

Fig. 2 Raman signal versus frequency. The *straight lines* denote the main observed peaks

the lattice disorder and the D′-peak around 2700 cm^{-1} which splits in two maxima. In the results, we can observe the dominant G-peak as in graphite bulk, the peak related to the disorder is not observable within the resolution providing us a hint for the good crystalline quality of the sample. The D′-peak shows similar behavior as in the literature when the sample is formed by a few layers graphene [9, 10].

Results and discussion

Figure 3 shows the temperature dependence of the measured resistance in the local and non-local configurations. The observed temperature dependence is similar to that reported for similar samples [8] when the electrodes are of non-magnetic materials. Following the reported systematic and for the thickness of the sample, we obtain a resistivity $125 \pm 25 \mu\Omega$ cm in agreement with reported data for similar samples [8]. However, from the measured two-points measurement, we obtain a much larger effective resistivity $\rho \simeq 870 \mu\Omega$ cm at 4 K mainly due to the contact resistance R_b formed at the interfaces Co/Pt/Graphite. From the measurement of these resistances in the two- and four-contacts configurations, we can estimate a contact resistance (Resistance × contact area) $R_b \sim 10^{-10}\,\Omega m^2$. Fert and Jaffrès [11] have shown that the realization of spin injection can be effective if the interface resistance R_b is larger than the product of the resistivity of (in the case) the graphite flake and the spin diffusion length (ℓ_s) in it. Assuming a similar spin diffusion length as the one measured in graphene $\ell_s \sim 1\,\mu m$ [12, 13], we obtain that $R_b > \rho \ell_s \sim 10^{-12}\,\Omega m^2$ and therefore, we may expect to see spin polarized transport signals in the graphite device.

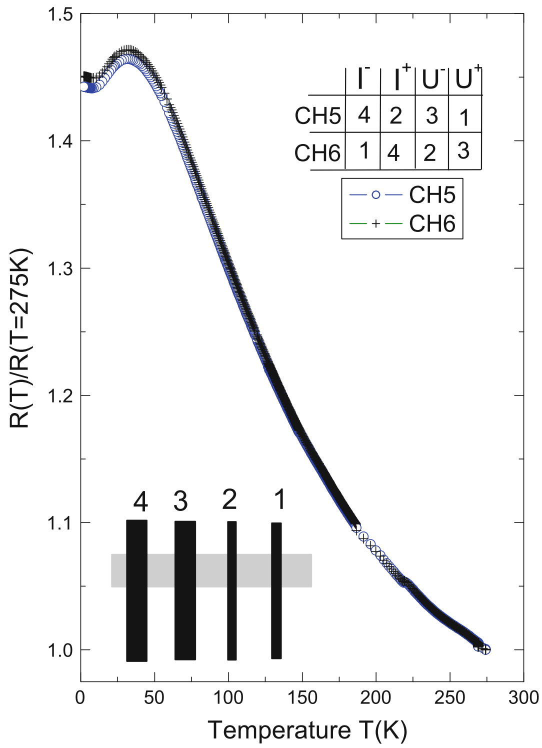

Fig. 3 Resistance normalized by its value at 275 K obtained for two configurations. *circle* configuration of channel 5 with the input current at electrodes 2 and 4, *plus* channel 6 with input current at electrodes 1 and 4 (see *inset* in the figure). The normalization resistance is $R(275) = 26\,\Omega$

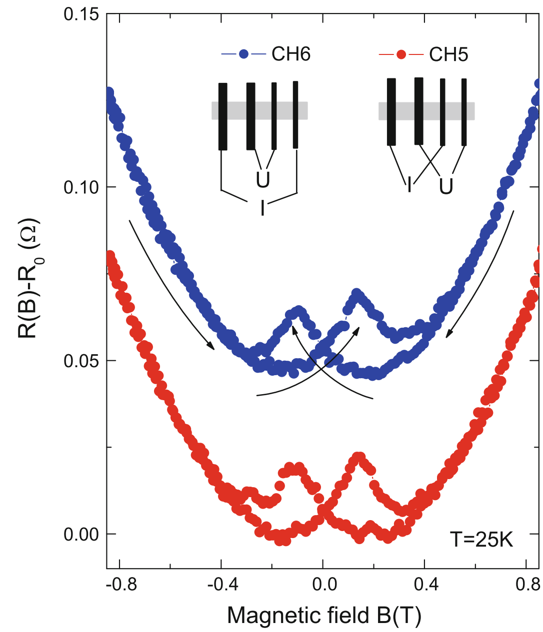

Fig. 4 Resistance of the two channels minus a constant arbitrary value R_0 versus applied magnetic field for the two configurations and at 25 K fixed temperature. The field was applied parallel to the main length of the Co electrodes and to the graphene layers

The measurements were done in a local and semi non-local configuration. The results presented in Fig. 4 were obtained at 25 K in both configurations as described in the figure. We clearly observe the two maxima at the opposite quadrants depending on the sweeping direction of the magnetic field. The maxima in the magnetoresistance appear within a parabolic dependence due to the unpolarized spin transport magnetoresistance of the graphite flake, this last probably due to a small misalignment of the sample. A positive (or sometimes a negative) contribution to the magnetoresistance as a consequence of a spin polarized transport is expected [14]. We note that in the case of spin transport in carbon nanotubes maxima [15] as well as minima [16] in the magnetoresistance were observed. In the device, the influence of the different widths of the ferromagnetic Co electrodes is manifested as a difference between the coercive fields. For example at 25 K the maximum at the positive field direction is located at $B_+ \sim 0.14\,\mathrm{T}$ whereas at the negative side $B_- \sim 0.1\,\mathrm{T}$.

Figure 5 shows the spin polarized magnetoresistance loops at different temperatures. These curves were obtained after subtracting the parabolic contribution of the non-polarized magnetoresistance at each temperature. The spin-dependent transport in the measurement and under the experimental conditions we used is observed up to 150 K. This temperature is relatively high taking into account that the spin injection is realized trough a metallic barrier (Pt) and not using an oxide barrier as usual. Other interesting point is the relatively long distance of $\sim 1\,\mu\mathrm{m}$ between the electrodes, see Fig. 1. This implies that the spin diffusion length should be larger than this length for $T < 150\,\mathrm{K}$. This value is similar to that observed in measurements at 4 K and at room temperature in graphene samples [17, 12] but larger than the one reported for a 2-nm thick graphite sample [6].

Because the coercive field of Co decreases with temperature also the field positions of the maxima as well as the resistance increase ΔR at the maxima decreases, see Fig. 5. Figure 6 shows the direct correspondence between the resistance height ΔR at the maxima versus temperature. In the same figure, we show the two coercive fields $B_{c\pm}$ (absolute values). The observed similarity in the temperature dependence indicate that the decrease in the spin dependent signal is related to the properties of the used ferromagnetic Co material and not due to the interface or graphite itself. Above ~ 200 K the observed maxima were overwhelmed by the measurement noise.

The results suggest that spin-dependent transport devices using tens of nanometer thick graphite flakes of micrometer size are possible. Further advantage of such a device is related to the relatively simple preparation method including the spin-injection/detection electrodes for which no extra dielectric barrier is necessary. This

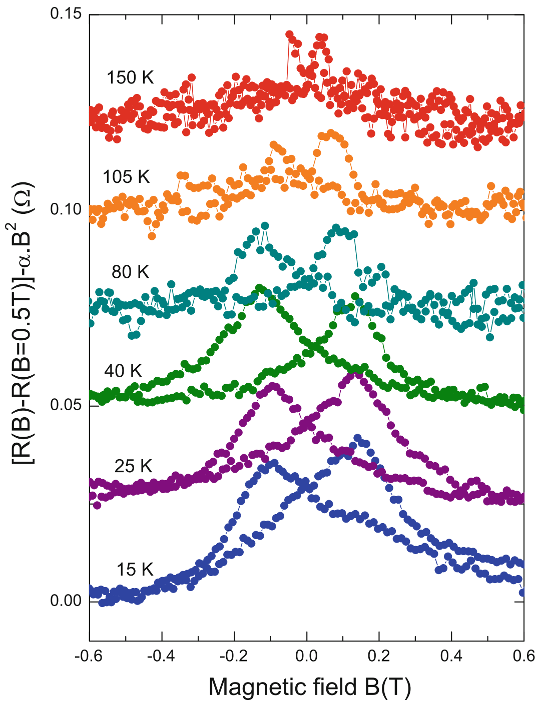

Fig. 5 As in Fig. 5 but at different temperatures for the local configuration only (channel 6)

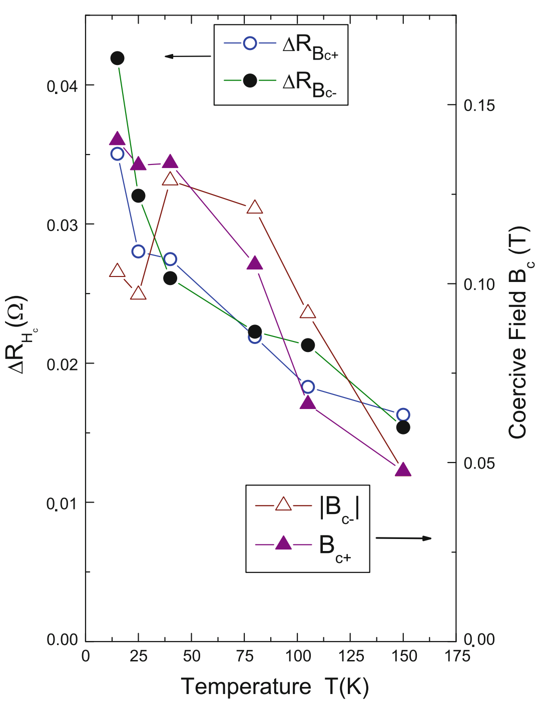

Fig. 6 *Left y* − axis: Resistance height at the maxima ΔR versus temperature in both sweep directions. *Right y* − axis: similar for the extracted coercive fields

reduces the contact resistance increasing the maximum current one may use in such a device. Note that for spin-torque experiments high spin currents are necessary. The difficulties to observe spin-dependent transport at temperatures higher than 150 K as well as the small relative change in the resistance may be overcome by using other spin injector materials or reducing the electrodes distance. Because the mobility of the graphite carriers increases the carrier density decreases [1], one may also try to use a gate control to increase the spin diffusion length.

Conclusion

In this study, we have experimentally shown that spin transport within micrometers in a 30-nm thick graphite flake is realizable up to a temperature of 150 K. We have also demonstrated that spin injection from a ferromagnetic Co electrode across a Pt thin layer, instead of the typical oxide barrier, is possible and may be useful for future applications.

Acknowledgements We gratefully acknowledge discussions with M. Ziese and U. Teschner for the Raman measurements. This study is supported by the DFG under DFG ES 86/16-1

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