Mater. Res. Soc. Symp. Proc. Vol. 1181 © 2009 Materials Research Society 1181-DD02-03

Tunneling and anisotropic-tunneling magnetoresistance in iron nanoconstrictions fabricated by focused-ion-beam

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ABSTRACT

We report the magnetoresistance (MR) measurements in a nanoconstriction fabricated by focused-ion-beam (FIB) in the tunneling regime of conductance. The resistance of the contact was controlled during the fabrication process, being stable in the metallic regime, near the conductance quantum, and under high vacuum conditions. The metallic contact was deteriorated when exposed to atmosphere, resulting in a conduction mechanism by tunneling. The TMR was found to be of 3% at 24 K. The anisotropic tunneling magnetoresistance (TAMR) was around 2% for low temperatures, with a field angle dependence more abrupt than in bulk Fe. This preliminary result is promising for the application of this technique to fabricate stable ferromagnetic constrictions near the atomic regime of conductance, where high MR values are expected.

INTRODUCTION

The mechanism of electronic transport in constrained geometries on the nanometer scale changes dramatically in comparison with bulk, once the dimensions are reduced to less than the mean free path of electrons. In this regime, electronic transport is not diffusive anymore, but ballistic, and the conductance can become quantized ($G=nG_0$, where $G_0=2e^2/h$ is the conductance quantum) [1]. In the case of magnetic materials, high-impact results were reported in the past in atomic-size contacts, with extremely large values for MR, phenomenon coined as "ballistic MR" (BMR) [2,3]. This effect was explained by the pinning of a domain wall in the constriction, restricting the transmission of electrons. However, subsequent experiments revealed that mechanical artifacts were playing a major role for these large ratios, resulting in a huge controversy [4]. Another different finding in these structures was a large anisotropy in the MR of the contacts, whose magnitude and angular dependence were found to be very distinct from bulk materials, the so called "ballistic anisotropic MR" (BAMR) [5,6,7]. This phenomenon seems to be better established than the BMR, although critical voices claim that atomic reconfigurations in the contact, rather than an intrinsic electronic effect, could explain this behavior [8]. For a recent review in the topic see reference 9. It is therefore crucial, for the study of these effects, the fabrication of stable nanocontacts, where mechanical artifacts are avoided. Most of the work in this field has been done by techniques such as scanning tunneling microscope, mechanical break junction (MBJ) and electrochemical junctions [1,9]. New nanolithography techniques such as FIB [10,11] and electron beam lithography [12,13] have been recently used to fabricate nanoconstrictions, opening an interesting new route in this field, since all the structure, including the contact, is attached to the substrate. Thus, mechanical artifacts are minimized, and the developing of devices based on these effects would be, in principle, feasible. However, the fabrication of constrictions by these techniques in the sub-100 nm range is extremely difficult, and the creation of atomic-size constrictions is especially challenging. We recently demonstrated a new way to fabricate stable atomic-size constrictions, by the control of the resistance while the FIB etching is performed [14,15]. In this work we show the first magnetoresistance results in one iron nanoconstriction obtained by this method.

EXPERIMENT

The constriction was fabricated at room temperature in a commercial dual-beam equipment (Nova 200 NanoLab from FEI). A previous optical lithography step was carried out before the etching process, patterning large 10 nm-thick Fe pads on top of SiO_2. The Ga-FIB etching was done in a region of around 2×6 μm^2 (red square in Fig. 1a), with a beam acceleration voltage of 5 kV and a beam current of 10 pA. Two electrical microprobes by Kleindiek were contacted on the pads (see Fig.1a). These conductive microprobes are connected via a feedthrough to a 6220 DC current source- 2182A nanovoltmeter combined Keithley system, located out of the chamber, measuring the resistance of iron *in situ* while the FIB etching is being performed. The etching process lasts a few minutes, roughly corresponding to a total ion dose of 10^{17} ions/cm^2. In figure 1b) the microstructure of the constriction after the milling is shown and in the inset the conduction channel before the etching process. For experimental details see reference 14.

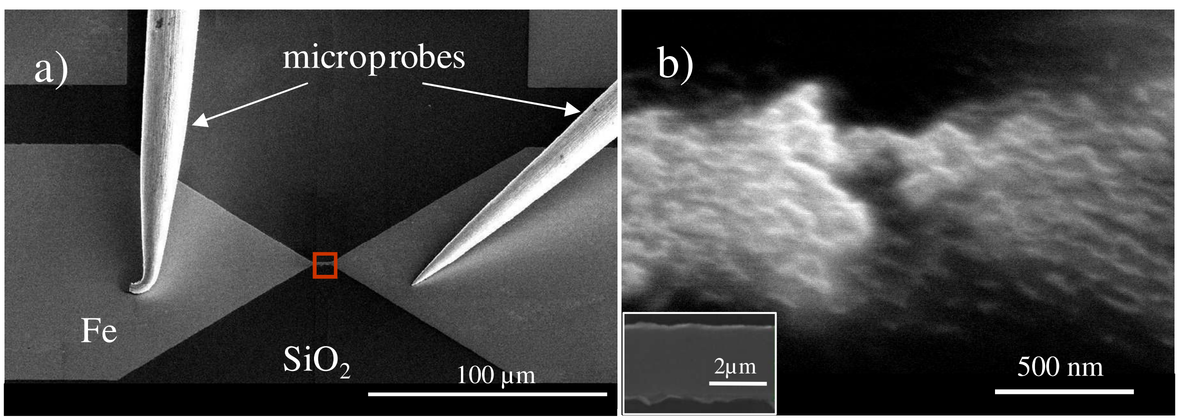

Figure 1. 52º tilted-view SEM images of the fabricated constriction. **a)** Experimental configuration, with the two microprobes contacted to the Fe pads for the *in situ* control of the resistance while the process is taking place. The red square indicates the etched zone. **b)** Microstructure of the iron after etching. A constriction is formed as a consequence of the ionic bombardment. The inset shows the metallic channel before the etching.

RESULTS

In figure 2 we show the evolution of the resistance with the process time. Since the initial resistance of the iron electrode is R_i ~3 kΩ, the resistance corresponding to the etched part (R_c) is the measured value minus R_i ($R=R_i + R_c$). After 2 minutes of etching, R starts to increase abruptly, and the FIB column is stopped when $R_c \approx 8$ kΩ. The R is measured during several minutes, finding that the constriction, in the metallic regime, is stable under the high vacuum conditions of the chamber (P~10^{-6} mbar). To avoid the possible deterioration when exposed to ambient conditions, a ~10 nm-thick layer of Pt-C was deposited by electrons (FEBID) on top of the etched zone, using $(CH_3)_3Pt(CpCH_3)$ as gas precursor. The high resistance of this material, because of the high amount of carbon, guarantees a resistance in parallel to the constriction of the order of tens of MΩ [16], which is confirmed by the negligible change of R while the deposit is done.

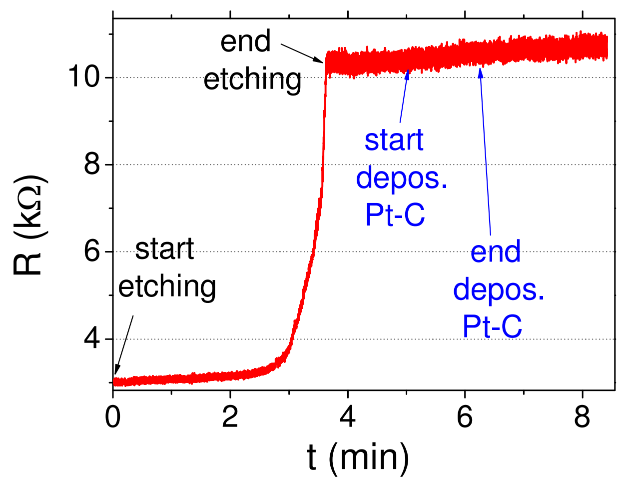

Figure 2. Resistance of the iron electrode as a function of the process time. The FIB etching is stopped when the resistance reaches 11 kΩ (R_c~8 kΩ). The constriction is covered by a thin layer of Pt-C, deposited by FEBID. The fabricated nanostructure is stable inside the vacuum chamber.

Once the constriction had been fabricated, the sample was put in contact with the atmosphere during some minutes, and transferred to a closed cycle refrigerator that can reach a minimum temperature of 24 K, combined with an electromagnet. The measurements of the resistance at room temperature showed an increase of the resistance by a factor of 10 (R≈100 kΩ), evidencing the departure from metallic conduction in the nanoconstriction. This value is significantly above the resistance corresponding to the quantum of conductance ($R_0=1/G_0=12.9$ kΩ). The slight non-linearity of the current-versus-voltage curves suggests that the constriction is not anymore in the metallic regime, but in the tunneling one (see Fig. 3a). The high reactivity of the nanostructure due to its large surface-to-volume ratio seems to be the most reasonable explanation of this effect. The resistance increased up to 120 kΩ ($G>9G_0$) at T=24 K.

We measured the MR for low temperatures with the magnetic field (H) parallel to the current path. In figure 3b the evolution with H and T is shown. MR ratios of the order of 3.2% are obtained. This value is a factor 30 times higher than in Fe non-etched samples, used as reference, where we observed a MR = -0.11 % in this configuration (note the different sign). As it is schematically explained by the red arrows in the graph, the evolution of R is understood by a change from a parallel (P) to an anti-parallel (AP) configuration of the ferromagnetic electrodes, separated by an insulator. More in detail, starting from saturation, a first continuous increase in R is observed, of the order of 0.8 %. This seems to be caused by a progressive rotation of the magnetization (M) in one of the electrodes. At around H=700 Oe, an abrupt jump of the resistance occurs (MR =2.5%), since the magnetization of one electrode switches its direction, and aligns almost AP to the M of the other electrode (intermediate state: IS). The MR becomes maximum at H=1 kOe, when the magnetization in both electrodes is AP. At H≈2 kOe, the M in the hard electrode also rotates, resulting again in a low-R state (P). As T increases, the IS, previously explained, disappears. IS is probably caused by the pinning of the M by some defects present in the soft electrode. Thus, the increase of the thermal energy favors the depinning of M. When the temperature was increased above 35 K, the nanoconstriction became degraded, likely due to the pass of the electrical current.

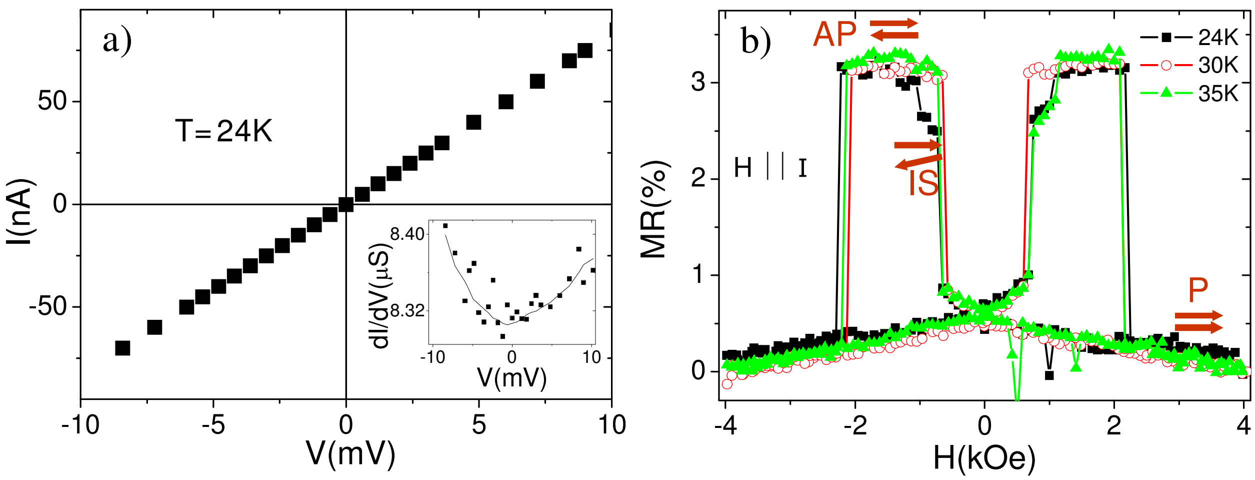

Figure 3. (a) Current-versus-voltage measurement at 24K. In the inset, the differential conductance of the curve deviates from the linear behavior, suggesting a non-metallic conduction. The line is a guide to the eye. **(b)** MR as a function of the magnetic field, applied parallel to the current direction. The MR is positive, with a value of 3.2% at 24 K, and can be understood by the decoupling of the magnetic electrodes separated by a tunneling barrier.

In figure 4a) the dependence of the MR at 24 K as a function of the voltage applied is shown, for the P and AP configurations. The diminishment of the resistance with the voltage is a typical feature of tunnel junctions, attributed to several factors such as the increase of the conductance with bias, excitation of magnons, or energy dependence of spin polarization due to band structure effects [17]. As the magnetostrictive state of parallel and antiparallel electrodes is the same, it seems that magnetostriction is not the cause of the observed TMR effect.

We have also studied the dependence of the MR with the field angle at saturation, by rotating the sample at the maximum field attained, H=11 kOe. In figure 1b) the evolution of MR is shown as a function of θ, the angle formed between H and the substrate plane. An anisotropic magnetoresistance effect (AMR) is present in the tunneling regime (TAMR). This effect is around 2% at 24 K, higher and of different sign from the AMR occurring in the bulk material (~ -0.3%). The TAMR has been previously observed in iron nanocontacts fabricated by MBJ [14], and implies that the evanescent wave functions maintain a strong atomic orbital character. The angle dependence is found to be more abrupt than the normal one for the AMR, proportional to $\cos^2\theta$. This behavior is typical for BAMR [5,14], and can be understood by considering the details of orbitals overlap [6,7].

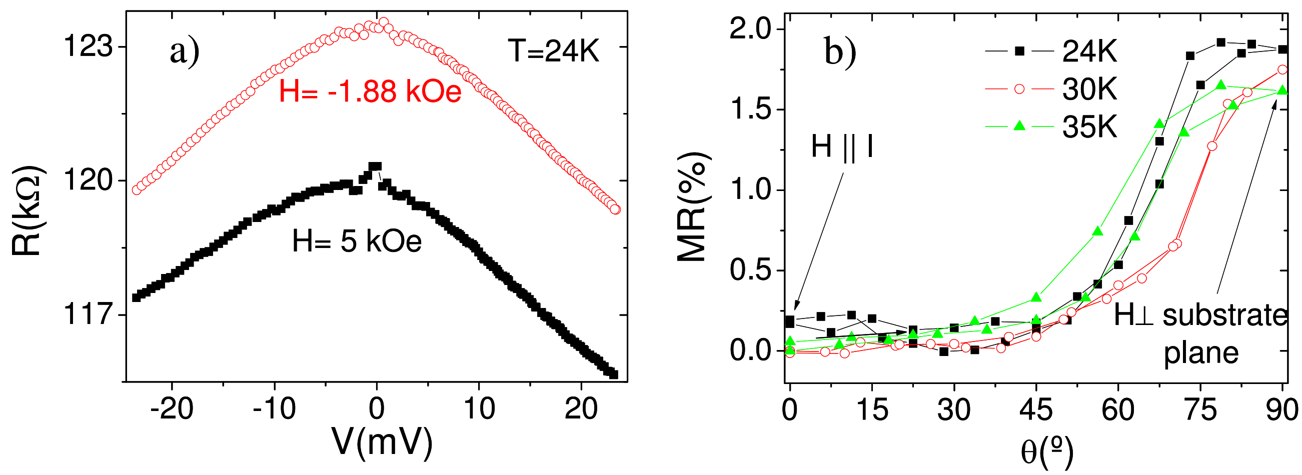

Figure 4. a) Bias dependence of the magnetoresistance at T= 24K for AP (H= -1.88 kOe) and P (H= 5 kOe) configurations. **b)** Angle dependence of the MR for low temperatures. A fixed field, H=11 kOe, is applied.

The experiments as a function of angle are also helpful to discard that magnetostriction effects are responsible of the observed MR. The magnetostriction in iron is given by

$$\frac{\Delta l}{l} = \frac{3}{2}\lambda_S(\cos^2\theta - \frac{1}{3}) \qquad (1)$$

with $\Delta l/l = -7 \times 10^{-6}$ for $\theta = 0º$, and $\Delta l/l = 3.5 \times 10^{-6}$ for $\theta = 90º$.

Thus, the increase of the MR as a function of θ has a contrary sign that if it was caused by magnetostriction, where the minimum resistance would be expected when H is perpendicular to the substrate plane.

CONCLUSIONS

In this contribution, we have shown the possibility to fabricate magnetic nanoconstrictions near the ballistic regime of conductance by the method we developed previously, using a FIB [14]. The constrictions are stable at room temperature and under high vacuum conditions. However, we have shown in this work the difficulty to have stable nanocontacts when exposed to ambient conditions.

An iron constriction in the tunneling regime, presents TMR ratios at low temperature 30 times higher than non-etched samples. We also observe a TAMR effect at low temperatures, of the order of 2%, and with an angle dependence different from the $\cos^2\theta$ expected for bulk AMR. From these experiments we also assure that magnetostriction is not playing any role in the measurements.

This result evidences that under low voltages etching, and with a moderated ion dose, the FIB procedure does not destroy the magnetic properties of the devices, although more research is required to investigate at what extent they are affected. The high stability expected for these constrictions in comparison with other suspended atomic-size structures, makes this first result promising for future research. A systematic study of the MR is currently under progress to improve the stabilization of constrictions in the metallic range of resistances, and investigate if high BMR and BAMR values can be attained at room temperature, which would have high impact in the field.

ACKNOWLEDGEMENTS

Financial support by the Spanish Monistry of Science (through project MAT2008-06567-C02, including FEDER funding), and the Aragon Regional Government are acknowledged.

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