

# Pseudo spin-valves with Al or Nb as spacer layer: GMR and search for spin switch behaviour

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**Abstract.** The magnetoresistance of several Ferromagnet/Normal metal/Ferromagnet spin-valve type structures has been investigated using Al as normal spacer layer. A magnetoresistance ratio up to 4.1% at room temperature and 5.7% at 0.3 K is found for the sandwich with both Co layers, while slightly lower signals are found for the structures involving CoFe and NiFe layers. The magnetoresistance dependence for Co/Al/Co, Co/Al/CoFe and Co/Al/NiFe on the spacer layer thickness exhibits the familiar non monotonic behaviour with second peak slightly larger than the one reported for Cu based pseudo spin valves. At cryogenic temperatures, preliminary results on the onset of spin switch effects in Co/Al/Co and the full spin switch effect in Co/Nb/Co are also reported here.

**PACS.** 72.25.-b Spin polarized transport – 85.75.-d Magnetoelectronics; spintronics: devices exploiting spin polarized transport or integrated magnetic fields – 73.43.Qt Magnetoresistance

## 1 Introduction

Since the discovery of the Giant Magnetoresistance (GMR) effect [1–3] the field of spintronics has gained a lot of interest for fundamental physics as well as applications. The applied side is based on the very rapid commercial success of giant magnetoresistance devices as magnetic field sensors in the read-heads of hard-disks [4,5], entering large-scale production within ten years from discovery. Moreover, magnetic random access memories (MRAM) based on Tunneling Magnetoresistance [6] have the potential of replacing CMOS based non-volatile FLASH memories in the future. For these devices to work, one needs to be able to precisely manipulate the dynamics of the spin in solid state devices.

This paper reports an experimental study on electrical spin dependent transport in sandwiches consisting of two ferromagnetic layers (F) separated by a non magnetic layer (N) involving magnetic materials such as Co, Co<sub>84</sub>Fe<sub>16</sub> and Ni<sub>80</sub>Fe<sub>20</sub>. The GMR signal of rf-sputter deposited F/N/F trilayers of the pseudo-spin-valve type was analysed. The resistance of the trilayer is expected to depend on the relative orientation of the magnetizations in the magnetic layers, with maximum GMR signal achieved in the antiparallel configuration. Antiparallel configuration can be achieved if magnetic materials have different coercive fields, i.e., different magnetization switching

fields. For this purpose, when the same magnetic material was used, bottom and top electrodes were deposited with different thickness, that allow to achieve different coercive fields. We used aluminum as spacer (normal) layer since it is a rather uncommon material for this purpose [7,8]; furthermore it can become superconducting at cryogenic temperatures making the trilayers suitable for investigating spin switch effects. The oscillatory GMR signal related to oscillatory coupling [9–12] as a function of spacer layer thickness is also reported here. The behaviour of the interlayer coupling as a function of the spacer thickness has been extensively studied in recent years [13] and it has been observed to follow the Ruderman-Kittel-Kasuya-Yosida (RKKY) coupling mechanism [14], while affected by some aliasing effect [15]. It is, by now, well established that the periods of oscillatory exchange coupling (OEC) are related to the Fermi surface of the spacer. This is particularly the case for Cu spacers for which this relation has been confirmed quantitatively [16–18]. In the past an experimental investigation [7] of OEC based on magnetic measurements (magnetisation) was reported for Fe/Al/Fe trilayers. Here we report an investigation based on electrical measurements (GMR signal) for trilayers involving different magnetic materials. At cryogenic temperatures, we also report the onset of a spin switch behaviour in a Co/Al/Co spin valve with rather thick Al spacer layer, and preliminary results on a Co/Nb/Co pseudo spin valve

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that exhibits full spin switch behaviour at liquid helium temperature.

## 2 GMR in pseudo spin valves with Al as normal spacer layer

Several F/N/F trilayers were fabricated to analyse the GMR signal. In particular series of Co/Al/Co, Co/Al/CoFe and Co/Al/Ni<sub>80</sub>Fe<sub>20</sub> were fabricated. The samples were prepared by RF magnetron sputtering in a high vacuum system with a base pressure of  $\cong 2 \times 10^{-7}$  Torr and were deposited in argon pressure at 3.1 mTorr at room temperature. The diameter for the magnetic and normal targets is 6 inch.

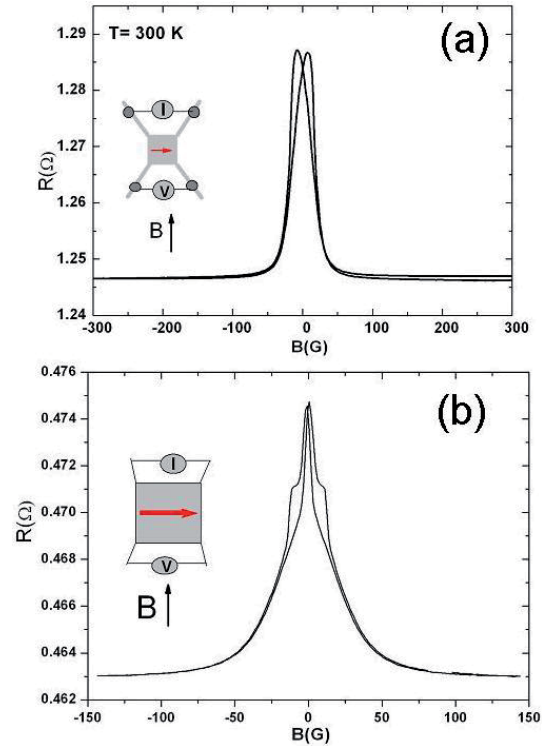
All the investigated trilayers have Co as bottom electrode, so we spent some time to find the best deposition parameters ensuring the minimum roughness. The Co roughness was found to generally decrease with used power and deposition rate. We found the optimum at a deposition rate of 0.04 nm/s, achieved using a power low as 100 W and with sample in intermittent motion under the 6 inch target. The spacer Al was deposited at 50 Watt with a rate of 0.18 nm/s achieved with sample stationary under the target.

The Co/Al/Co trilayers were deposited onto a  $10 \times 10$  mm<sup>2</sup> glass substrate in an applied magnetic field  $B_{\text{growth}} = 1000$  G, to induce an easy axis for magnetization. The bottom Co layer is 8 nm thick while the top Co layer is 16 nm thick, and the trilayer is covered with  $\cong 2$  nm Al layer to prevent the oxidation of the top layer. From an AFM analysis a roughness smaller than 0.5 nm was found for our Co, thus the thickness of deposited Al spacer layer must be larger than this value. In Figure 1a we show the GMR signal for a trilayer with Al = 3.6 nm. Data were recorded at room temperature applying an in-plane magnetic field transversal to the current direction, and the resistance of the sample was measured as sketched in the inset. The whole trilayer was patterned by photolithography and wet etch in a square with 5 mm sides and with fingers 0.2 mm wide contacting the edges of the square.

The magnetoresistance  $\Delta R/R$  is defined as the maximum change in resistance observed over the field range of interest divided by the high field resistance. From data  $\Delta R/R \cong 4\%$ .

Unpatterned Co/Al/Ni<sub>80</sub>Fe<sub>20</sub> structures were fabricated with the same process as for the Co/Al/Co trilayer. The Co layer is 8 nm thick and the permalloy is 8 nm thick, topped by a 3 nm Al cap layer. In Figure 1b the GMR signal of a trilayer with spacer layer Al = 3.6 nm is plotted. A GMR signal of 2.2% is found for this structure.

We also investigated unpatterned Co/Al/CoFe trilayers. The structure was sputter deposited onto an insulating Si (100) substrate in applied magnetic field,  $B_{\text{growth}} = 1000$  G. The Co layer is 12 nm thick and the Co<sub>84</sub>Fe<sub>16</sub> is 14 nm thick. A 3 nm thick Al film was used as cap layer. CoFe is deposited at 150 Watt at a rate of 0.1 nm/s. Maximum GMR signal of only 1.1% was found for this last structure.



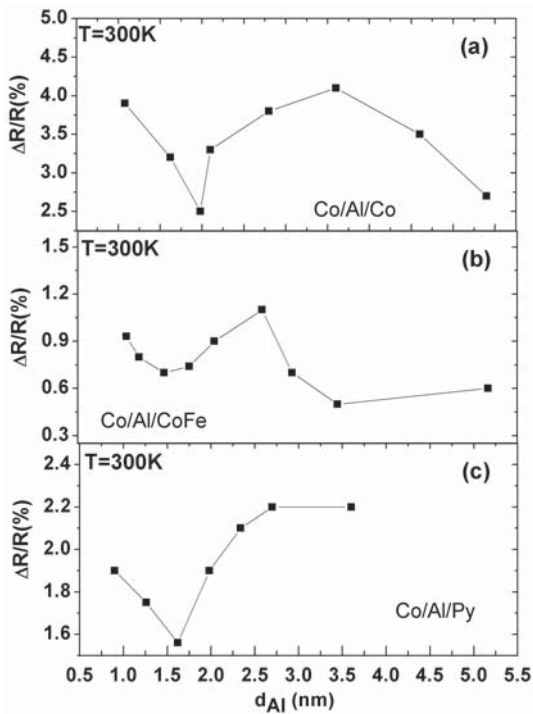
**Fig. 1.** Room temperature resistance versus field curve for (a) Co(8 nm)/Al(3.6 nm)/Co(16 nm) trilayer and (b) Co(8 nm)/Al(3.6 nm)/Ni<sub>80</sub>Fe<sub>20</sub> (8 nm) trilayer, The samples are grown in applied magnetic field,  $B_{\text{growth}} = 1000$  G, to induce an easy axis for magnetization (arrow on the chip).

It must be said that although the GMR signals for all the trilayers we report in this work are not large (but consistent with Cu based pseudo spin valves, i.e., few percents), yet they are larger than the AMR signal, (not shown here) of the single magnetic layers, all well below 1%. That is why we are dealing with a GMR signal.

Three series of trilayers with variable Al thickness were fabricated to study the GMR behaviour as a function of the Al spacer layer. Al thickness for the Co/Al/Co and Co/Al/CoFe sandwiches varies from 0.9 nm to 5.4 nm while in Co/Al/NiFe the range is 0.9–3.6 nm. Each point in the plot in Figure 2 was extracted from the MR curve obtained applying a magnetic field transversal to both the current and the easy axis for magnetization.

For all the three series, as shown in Figure 2, the magnitude of magnetoresistance oscillates with increasing Al layer thickness. Figure 2a shows an oscillating magnetoresistance with maximum peak 4.1% at  $d_{\text{Al}} = 3.6$  nm. Slightly lower magnetoresistance peaks were found for the other two trilayers, see Figures 2b and 2c. The higher signals in the case of the Co/Al/Co trilayers comes from the better roughness of their Co layers. We should expect the magnetoresistance to increase as  $d_{\text{Al}}$  decreases, unfortunately we could not allow Al thinner than 0.9 nm because of the Co bottom layer roughness.

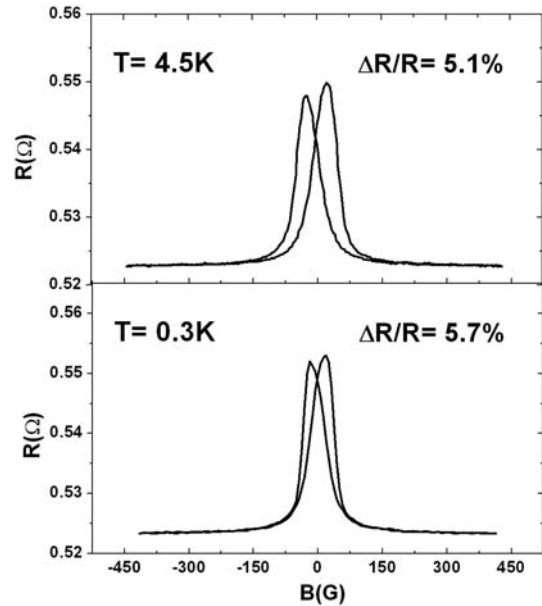
The plots in Figure 2 cannot provide information about the first peak of the oscillating curve since it would



**Fig. 2.** Transverse magnetoresistance vs. Al layer thickness for (a) glass/Co(8 nm)/Al( $d_{Al}$ )/Co(16 nm), (b) Si(100)/Co(12 nm)/Al( $d_{Al}$ )/Co<sub>84</sub>Fe<sub>16</sub>(14 nm), (c) glass/Co(8 nm)/Al( $d_{Al}$ )/Ni<sub>80</sub>Fe<sub>20</sub>(8 nm) taken at room temperature. The line is a guide for eyes.

show up for lower Al thickness. We observe that the second peak is found for Al thickness around 2.5 nm for Co/Al/CoFe trilayers, while for the Co/Al/Co and Co/Al/Py structures it occurs at a slightly larger value,  $\approx 3.5$  nm. Copper based heterostructures have been extensively studied in the past [19,20]. For comparison, Parkin et al. [11] in their experiments on antiferromagnetically coupled Co/Cu superlattices found that the second peak of the GMR curves as function of the Cu spacer thickness was  $\approx 2.5$  nm and Marrows [21]  $\approx 2.2$  nm. Thus, our Co/Al based structures give a peak comparable with the ones observed in the past experiments on Cu based structures, though slightly larger. This, in the framework of RKKY model with aliasing effect [14,15], should be expected because of the larger lattice constant of Al with respect to the Cu. Moreover, the range of Al thickness for appearance of ferromagnetic (lower MR) or antiferromagnetic (larger MR) coupling between electrodes magnetisations is found in reasonable agreement with the one extracted from magnetic measurements [7] on Fe/Al/Fe structures.

Finally, we recorded the magnetoresistance of the trilayers at cryogenic temperatures. As expected, the GMR was found to increase appreciably. As an example, in Figure 3 we show the MR curve recorded at 4.5 K and 0.3 K for the Co/Al(3.6 nm)/Co. As it is seen, GMR increases by 30–50% with respect to room temperature value (4.1%, see Fig. 2).



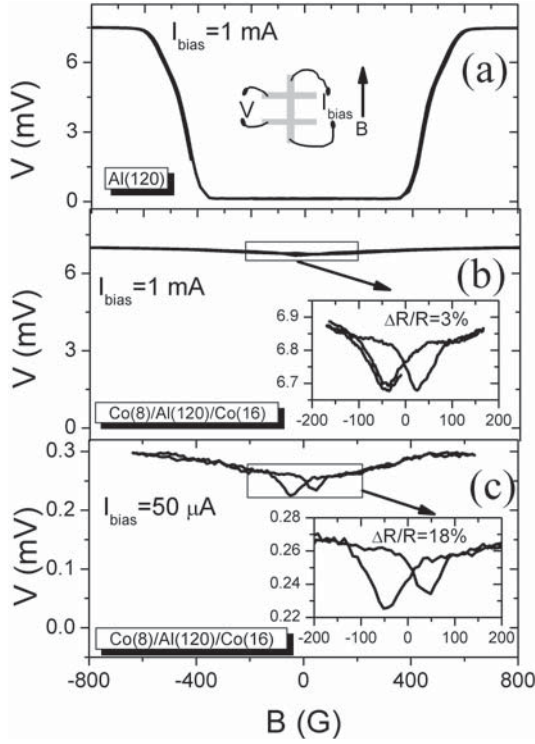
**Fig. 3.** Transverse magnetoresistance for glass/Co(8 nm)/Al(3.6 nm)/Co(16 nm) pseudo spin valve, at cryogenic temperatures.

### 3 Search for spin switch effects in Co/Al/Co and Co/Nb/Co pseudo spin valves

Below 1.2 K the Al films can be superconductive, and the spin valve would turn into an FSF sandwich exhibiting spin switch behaviour.

In a FSF spin switch, superconductivity can be controlled by the relative orientation of the magnetisations of the outer ferromagnetic electrodes sandwiching the superconductor. The possibility to control superconductivity by mean of a relatively weak magnetic field in such a way was proposed long time ago by de Gennes [22]. In the pioneering experiments the superconductivity was found to be depressed for parallel alignment of magnetisations of outer ferromagnetic insulators [23] or weakly coupled (a very thin insulating barrier at SF interfaces) metallic ferromagnets [24].

Recently spin switches based on proximity coupled metallic ferromagnets [25,26] and classic metal superconductors (as Nb) [27–32] or high  $T_c$  superconductors (as YBCO) [33] are under intense study. Experimental results reported until now suggest that superconductivity can be depressed both in the antiparallel state of the magnetisations (inverse spin switch effect) and in the parallel state (standard spin switch effect). As a general trend, standard spin switch effect is observed when exchange biasing is used [27–30] to achieve the antiparallel state, while the inverse effect is observed when antiparallel orientation is achieved using different coercive fields [31–33], as we made above in our pseudo spin valves. In the inverse spin switch effect many mechanisms can have some role, as non homogeneous superconductivity induced by domain walls [31], spin imbalance-induced depression of superconductivity [31–34], or more exotic mechanisms as

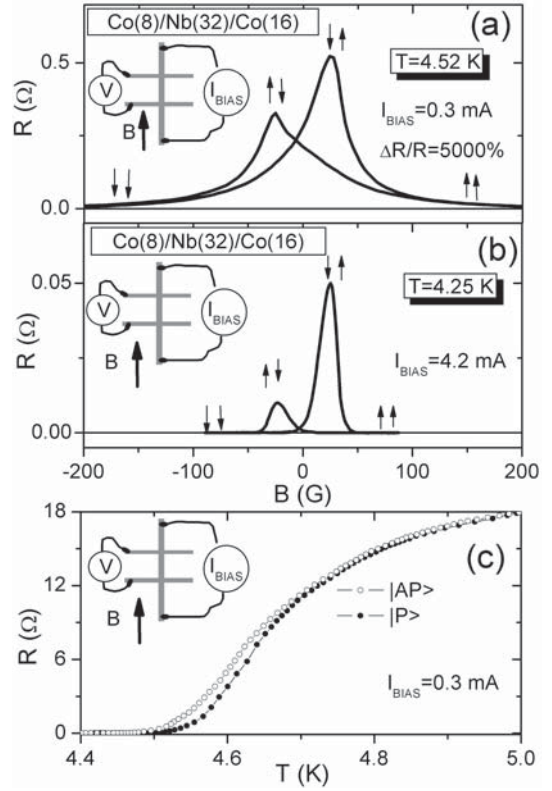


**Fig. 4.** Voltage at constant current versus magnetic field for a 120 nm thick Al film (a) and for a glass/Co(8 nm)/Al(120 nm)/Co(16 nm) trilayer biased with 1 mA (b) or 50  $\mu$ A (c).

inverse proximity effect or induction of triplet component of superconductivity [35].

In the FSF spin switch, superconductivity is observed at finite temperature only above a critical thickness for the S layer, (here Al). For the Al thickness used in the above discussion we never found superconductivity in Al above 0.3 K. In our tests for approaching from below the critical thickness of Al, we finally found onset of superconductivity at 0.260 K in a Co/Al/Co trilayer with 120 nm thick Al. We hence only can say that in FSF spin switch based on strong Ferromagnets (as our Co, NiFe, CoFe) the critical thickness of Al is slightly larger than 120 nm at working temperatures above 0.260 K. Data recorded at 0.260 K for our glass/Co(8 nm)/Al(120 nm)/Co(16 nm) trilayer with Al near the critical thickness as well as a single Al layer of same thickness are shown in Figure 4. Both single Al layer and spin switch trilayer were patterned by photolithography and lift off for four contact measurements as shown in Figure 4a. The strip was 0.2 mm wide and voltage contacts were 5 mm apart. The magnetic field was applied in the plane of the strip and longitudinal to the current.

Figure 4a shows the voltage at constant current (proportional to the resistance) versus in plane magnetic field for the single Al film. As we can see, the superconducting critical field of the used Al film is about 600 G, with onset of transition to the normal state around 400 G. In Figures 4b and 4c voltage versus field of the trilayer is reported for two different biasing currents. For both biasing currents a spin dependent signal can be envisaged at



**Fig. 5.** Resistance at constant current versus magnetic field for a glass/Co(8 nm)/Nb(32 nm)/Co(16 nm) trilayer at 4.52 K (a) or 4.25 K (b). (c) Resistance versus temperature for the glass/Co(8 nm)/Nb(32 nm)/Co(16 nm) trilayer with applied magnetic fields promoting Parallel (solid circles) or Antiparallel (open circles) magnetisations in the ferromagnetic films.

low fields (below 100 G) superimposed to the background signal typical of magnetic field induced superconducting-normal transition of the Al film. The MR of the spin-dependent signal is much larger than the one expected from GMR effect, that for the used Al thickness we found of the order of 0.1%. The much larger MR suggest that the spin switch mechanisms is going to work, particularly for the lower bias current shown in Figure 4c were a MR as large as 18% is recovered. This is coherent with the fact that at lower bias currents we are nearer to the superconducting regime of Al film and so nearer to the spin switch effect that would be fully manifested if the Al film was slightly thicker.

We should remark that, in the classification given above, data in Figures 4a and 4b seem to suggest the onset of a standard spin switch effect in our Al based FSF pseudo spin valve, i.e., superconductivity is depressed for parallel orientation of the magnetisations in the outer electrodes. We stress that these are preliminary data on one sample, but, if confirmed by further measurements we are going to make in the next future, the pseudo spin valve Co/Al/Co could be an example where standard spin switch behaviour is observed without the use of exchange biasing to set the antiparallel state. We should notice that earlier observations [24] of standard spin switch effects in

pseudo spin valves, though with not clean interfaces, were made using a superconductor of the first type, as our Al.

To make more clear the discussion on our Al based spin switch, we anticipate here some new results on our recent investigations on a spin switch based on a second type superconductor, Nb, that indeed shows inverse spin switch effect, in agreement with recent results on Nb-based spin switches not using exchange biasing [31,32].

The structure is a trilayer Co(8 nm)/Nb(32 nm)/Co(16 nm)/Al(2 nm) deposited on glass and patterned as the Al-based trilayer shown in Figure 4. Due to the higher transition temperature of Nb with respect to Al, the whole superconducting regime is accessible for Nb above 4.2 K for Nb films thicker than about 30 nm.

In Figure 5a we show the MR of the trilayer at a temperature (4.52 K) slightly larger than the critical temperature (zero resistance temperature) of the trilayer. The magnetic field is directed longitudinal to the current, as shown in the inset. As it is seen, a MR ratio large as  $\Delta R/R = 5000\%$  is observed using a biasing current of about 0.3 mA. In Figure 5b we are at 4.25 K and we use a biasing current of 4.2 mA. Now the full superconducting regime ( $R = 0$ ) is achieved and resistive-superconductive transition is controlled by rather weak magnetic fields, below 100 G. These fields are much lower than parallel critical field of the used Nb films, estimated to be much larger than 2000 G at 4.2 K. As shown by the arrows in the insets, we expect the relative orientation for magnetisation of the outer ferromagnetic electrodes to be parallel at high fields and almost antiparallel for fields around +30 G and -30 G. So, the antiparallel state is detrimental to superconductivity in Co/Nb/Co structures, i.e., inverse spin switch effect is observed in this structure. Magnetoresistance curves in Figures 5a and 5b are slightly asymmetric, as if a small exchange biasing effect were working. This is possibly due to the fact that we recorded data after many thermal cycles that could have deteriorated the very thin (2 nm) Al cap layer. So, the presence of a thermally grown antiferromagnetic CoO on the surface of the top Co cannot be excluded. This thin CoO layer can account for a weak exchange biasing effect present in data. Finally, in Figure 5c we show the resistance versus temperature curve taken at a constant current of 0.3 mA for two fixed magnetic fields that promote an antiparallel state (|AP>) or a parallel (|P>) state for magnetizations. As measured in the middle of transition, a difference in temperatures of as about 30 mK between the parallel state and the antiparallel state  $R(T)$  curves is recovered. This difference in temperatures is of the same order of magnitude of the ones observed in proximity coupled FSF structures using Nb and strong magnetic materials [29–32] other than our elemental Co.

We notice that the biasing current was the same in Figures 5a and 5c. As it is known, the zero voltage state at a given temperature depends on the magnitude of the used bias current. As we will show elsewhere [36], and as it is intuitive, to the parallel and antiparallel states correspond two superconducting critical currents (extension in current of the zero voltage state). In the case of

the present spin switch, at a given temperature the critical current of the parallel state is larger than the critical current of the antiparallel state. In Figure 5b we used a bias current (4.2 mA) in between the critical currents at 4.25 K, so achieving a resistive state also at a temperature that from Figure 5c (acquired using a lower bias current, so exhibiting larger critical temperature) seems below the critical temperature of the trilayer.

## 4 Summary

We have found that the GMR signal in rf-sputter deposited sandwiches with Co, CoFe and NiFe electrodes and Al spacer layer is few percent; in particular, the best value was found for the Co/Al/Co trilayer,  $\Delta R/R = 4.1\%$  at room temperature and 5.7% at 0.3 K, possibly due to the deposition process which yielded smoother film surfaces. However, for all the fabricated trilayers the GMR signal is larger than the AMR signal of the single magnetic layers, so that we can assume that a GMR signal was always present. Furthermore, a qualitative analysis of the GMR signal in Co/Al/Co, Co/Al/Co<sub>84</sub>Fe<sub>16</sub> and Co/Al/Ni<sub>80</sub>Fe<sub>20</sub> varying the spacer layer thickness has also been carried out. As expected, the dependence of the GMR signal on the Al spacer thickness has an oscillating behaviour. The second peak of MR as a function of the Al spacer layer gave a result in qualitative agreement with the previous experiments on Co/Cu structures, with a slightly larger value for our Co/Al structures.

Tests of our trilayers at cryogenic temperatures show that Al is not superconducting above 0.3 K for thickness below 120 nm when interfaced with strong ferromagnets. We recorded an onset of superconductive transition with relative onset of spin switch behaviour, exhibiting a  $\Delta R/R = 18\%$  in fields below 100 G at 0.260 K, in a Co/Al/Co trilayer with 120 nm thick Al. So, in an Al based spin switch operated above 0.260 K a thickness larger than 120 nm should be used. Moreover, we achieved full superconductive regime with full spin switch behaviour in a Co/Nb/Co pseudo spin valve with 32 nm thick Nb at liquid helium temperature. Near the superconductive transition a  $\Delta R/R = 5000\%$  in fields below 100 G are recovered for this kind of spin switch. Though preliminary, results reported here seem to suggest a standard spin switch effect in the Al based pseudo spin valves, while Nb based spin valves exhibit an inverse spin switch behaviour.

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