Magnetotransport Properties and Tunnel Effect of Thin Film Nano-Structures

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ABSTRACT

Magnetotransport and tunnel effect measurements were made on nanostructured thin films. Nano-structures as FM/NM/FM/AFM were deposited by magnetron-sputtering on the oxidized Si wafers, as continuous films and cross-stripe structures. FM (Ferro-Magnetic) denotes NiFe (Py), Co or a combination using Py-Co layers. NM (Non-Magnetic) denotes Cu or Al_2O_3 layers. AFM (Anti-Ferro-Magnetic) denotes FeMn layers. These nano-structures present anisotropic magnetoresistance (AMR) and tunnel magnetoresistance (TMR) effects. On the unpatterned films we made magnetoresistance (MR) and Hall effect measurements in order to study the magnetotransport properties of the samples and surface quality. The effective thicknesses of the oxide layers were estimated by tunnel effect measurements and compared with in situ quartz microbalance measurements.

Keywords: thin films, anisotropic magnetoresistance effect, tunneling effect, Hall effect, microstructure

1 INTRODUCTION

The investigation of the electrical conductivity and magnetoresistance of magnetic multilayers and granular films plays an important role in the design and fabrication process of magnetic micro and nanosystems for spintronics. The aim of this work is to study some multilayered nanosystems and to present a fast and reliable method to characterize the electrical and magnetic properties of these structures. Using magnetoresistance (MR), Hall Effect and Tunneling Effect we can estimate the quality and other properties of the deposited films. Due to low pressure of the Ar gas and the presence of a small amount of O_2 in the deposition chamber, our samples obtained by sputtering method present intermixed regions of FM and NM layers. These intermixed regions are responsible for the increasing of the Hall resistivity which can be very attractive for magnetic sensors. The saturation fields obtained when the magnetic field is applied perpendicular to the film plane are less than the values predicted from the shape anisotropy and give us information regarding the films roughness. The extraordinary Hall Effect can be used as a simple and sensitive tool for the study of the magnetic properties of thin films and can lead to a new generation of magnetic sensors and memory devices.

2 EXPERIMENTAL

The samples investigated were prepared by r.f. magnetron sputtering into a ER 3119 CRYO VARIAN equipment. The base pressure was 5×10^{-7} torr. The Ar pressure during the deposition process was 5×10^{-3} torr and the rate of deposition was 0,01 nm/s. The samples, deposited onto oxidized Si wafers processed at laser quality, were denoted by S_1, S_2, S_3 and S_4. The structure of S_1 is Si/SiO_2/Py(17.1nm)/Co(3.3 nm)/Al_2O_3(1.3 nm)/Co(3.3 nm)/Py(17.1 nm)/FeMn(45 nm)/Py(8.6 nm), the structure of S_2 is Si/SiO_2/Py(17.1nm)/Co(3.3 nm)/Cu(2 nm)/Co(3.3 nm)/Py(17.1 nm)/FeMn(45 nm)/Py(8.6 nm) while for S_3 the structure is Si/SiO_2/Py(17.1 nm)/Al_2O_3(1.3 nm)/Py(17.1 nm)/FeMn(45 nm)/Py(8.6 nm) and for S_4 the structure is Si/SiO_2/Py(21 nm)/Al_2O_3(2 nm)/Py(21 nm).

The structures were obtained both as continuous (unpatterned) films and as tunnel junctions (samples $S_{1,3,4}$) by using shadow masks to create a cross-geometry structure of area $2.25\cdot10^{-6}$ m^2 as shown in figure 2a. The insulating barrier Al_2O_3 was grown by natural oxidization in air of the Al layer at room temperature for 36 h. For electrical conductivity, Hall Effect and tunnel effect measurements we used the dc four point technique. All these configurations are connected through a switching box to a Keithley system composed by a current source 6221 and a nanovoltmeter 2182A. Additionally, two instrumentation amplifiers (EI-1040) and an external DAQ board (Lab Jack U12) were used for test measurements.

3 RESULTS AND DISCUSSION

Figure 1 shows the AFM topography of the sample S_3. The maximum roughness of the film surface is about 5.69 nm. We can see sharp columnar grains that are growing perpendicular to the film surface. This value of the roughness and the surface aspect suggest the existence of intermixing effects between the adjacent FM and NM layers which will produce nonmagnetic layers at the interfaces. These factors will affect the effective thickness of the Al_2O_3 layers, the barrier height and the amplitude of the tunnel magnetoresistance effect (TMR). The thinner intermixing layer we have, the greater GMR and TMR value we can expect.

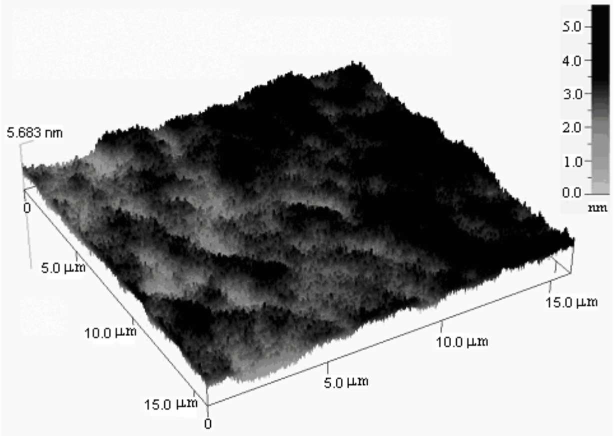

Figure 1: The AFM image of the surface for sample S_3

Figure 2 shows the experimental setup and the results of tunnel effect measurements made on samples $S_{1,\ 3,\ 4}$. The current-voltage data at room temperature were fitted by Simmons' theory of tunneling [1] to obtain values of tunnel barrier height (F) and the effective thickness (s) of the oxide layer. Excepting sample S_1 the experimental data and the fitting curves show typical behavior for a tunnel junction with metallic electrodes. The estimated thickness of the oxide layer, measured with the quartz microbalance during the deposition process, is smaller, for each sample, than the value obtained from tunnel experiments. For sample S_1 we obtained, as shown in Figure 2a, an almost linear characteristic with a high value, F =7.33 eV, of the tunnel barrier height and an effective thickness of about 7.41 nm. The values for F that we obtained for samples S_3 and S_4 are slight smaller then the values reported for FM/Al_2O_3/FM junctions. On the other hand, the thicknesses that we obtained from tunnel measurements for the oxide layers, s=4.74 nm compared with 1.3 nm for S_3 and s=7.18 nm compared with 2 nm for S_4 as presented in Figure 2b and Figure 2c respectively, show us that the adjacent permalloy layers are partially oxidized [2]. Good insulators like Al_2O_3 lead to F in the eV range whereas the magnetic oxides have F of fractions of eV [3] and lead to a decreases of the measured barrier height. The value for F that we obtained for sample S_3 (about 1.16 eV) is slightly smaller than the value reported for FM/Al_2O_3/FM which is about 1.6 eV. The barrier height for sample S_4 was found to be F =0.54 eV. The value of resistance of the sample S_1 is about 19.7 kO and suggests a high degree of oxidation of the metallic layers. It was shown in [2] that at least 2 nm from the metallic layer deposited on to substrate is oxidized due to SiO_2 layer. On the other hand we can have collision intermixing between the NiFe layer and the Co layer. In addition, during the growth of the Al_2O_3 layer, the metallic layer (Co) is being oxidized. In the same way, we can assume that at least 2 nm from the top layer is also oxidized. Because of these reasons the structure S_1 presents a high resistance. From the behavior illustrated in Figure 2a, we can assume that the conduction in this structure is made by electron tunneling between metallic grains which are surrounded by thin oxide barriers [4]. These facts explain the high values obtained for the barrier height and width. In other words, our measurements reveal an oxidized granular structure, slightly bellow the percolation threshold. We obtained an activation energy for conduction of about 1.6 eV.

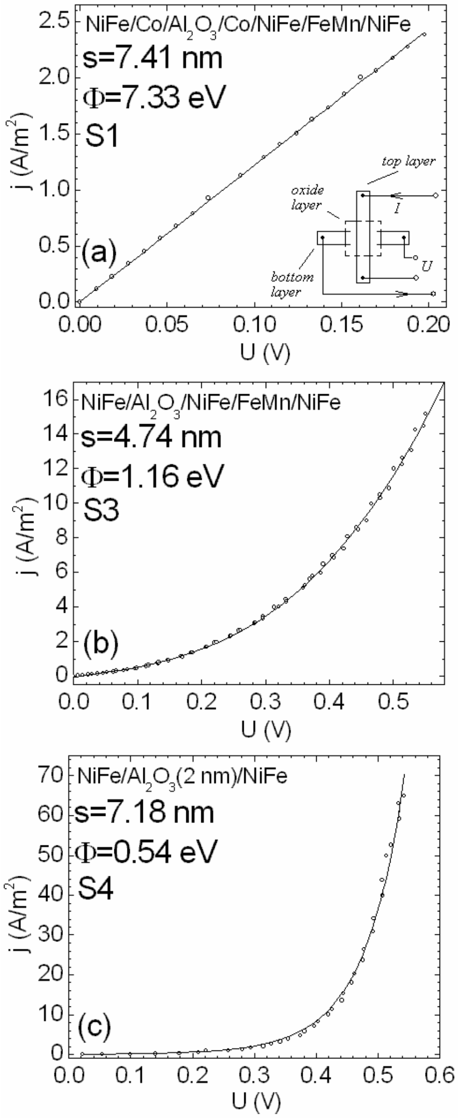

Figure 2: Results of the tunneling effect measurements made on (a) sample S_1, (b) sample S_3 and (c) sample S_4.

The replacement of the Al_2O_3 layer with a Cu layer, in sample S_2, leads to a decrease of about 7 times of the film resistance. The I-U characteristic of the sample is linear. However, the resistance of the sample is still high and we believe that the first two layers (NiFe/Co) are oxidized. The top layers are partially oxidized too. The temperature dependence of the resistance has the same aspect like for S_1 but the activation energy is much lower, about 0.27 eV.

For the sample S_3 we obtained a tunnel magnetoresistance effect (TMR) of about 0.5% with the magnetic field applied perpendicular to the top layer but in the film plane. The field dependence of the TMR effect for this sample is presented in Figure 3.

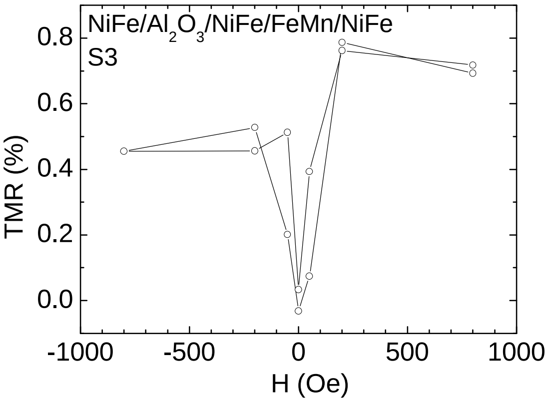

Figure 3: The field dependence of the TMR effect for the sample S_3.

However, because the bottom layer is almost oxidized, we believe that the observed effect is in fact due to anisotropic magnetoresistance effect (AMR) that take place in the top layer. The Hall Effect measurements were performed on the unpatterned films. We obtained a response in magnetic field only for samples S_2 and S_3. Figure 4 shows the field dependencies of the extraordinary and planar Hall Effect (PHE) in samples S_2 and S_3.

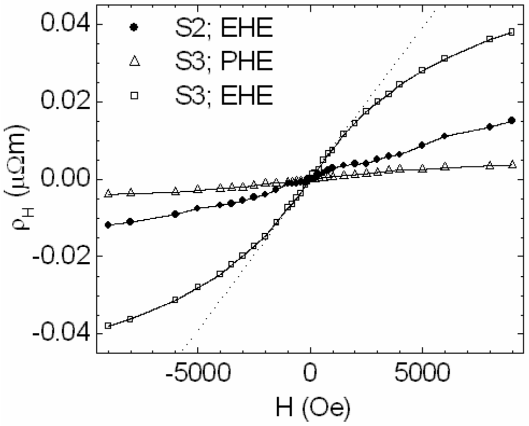

Figure 4: The field dependencies of the Hall resistivity for samples S_2 and S_3; EHE means the extraordinary Hall Effect and PHE the planar Hall Effect.

The high value of the extraordinary Hall resistivity, ?$_H$, for sample S_3 reveals a structure which behaves like a ferromagnet-insulator mixture in the vicinity of the percolation threshold [5, 6]. This result is confirmed by the value of the activation energy for conduction E_a=0.91 eV which is lower than the activation energy for sample S_1, that is almost completely oxidized, and higher than the activation energy for sample S_2, which has Cu as interlayer. The unpatterned structure, S_3, presents the giant Hall Effect which is very attractive for magnetic field sensors. The planar Hall Effect is very small for sample S_3 and virtually absent for sample S_2. The absence of the Co layer for sample S_3 permits to saturate more easily. The Cu layer has a shunting effect and lowers the value of the Hall resistivity.

Now, we can compare these values of the Hall resistivity with other measurements made on NiFe (10 nm) and NiFe (2 nm)/Al_2O_3 (1 nm)/NiFe (2 nm) prepared by thermal evaporation and presented elsewhere [7]. For the NiFe sample the magnetic phase is well defined and the PHE effect is relatively high. The sample saturates at a magnetic field which is less than the value predicted from the shape anisotropy and presents hysteresis at low fields [7]. However, the values of the Hall resistivity are about 50 times smaller than the values obtained for sample S_3. Figure 5 shows, for comparison, the field dependencies of the Hall resistivity for NiFe (10 nm) and NiFe (2 nm)/Al_2O_3 (1 nm)/NiFe (2 nm) thin films.

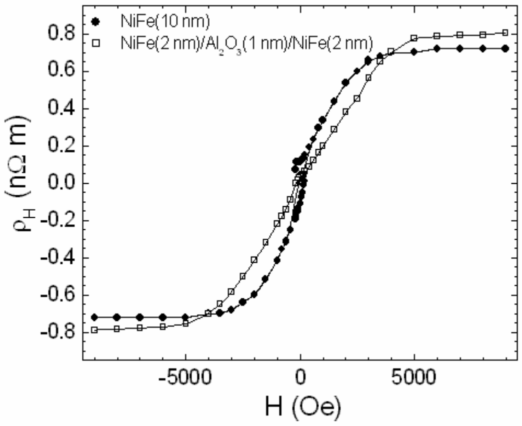

Figure 5: Field dependencies of ?$_H$ for Py(10 nm) and Py(2 nm)/Al_2O_3(1 nm)/Py(2 nm) thin films (Py denotes NiFe alloy) deposited by thermal evaporation on oxidized Si substrates.

The decrease of the saturation fields is related with the surface quality. This is the roughness effect [8]. For a 10 nm thin film of permalloy obtained by thermal deposition the saturation field decreases to about 4000 Oe. This leads to a roughness effect about 8 nm, which is in good agreement with the AFM measurements. For NiFe (2 nm)/Al_2O_3 (1 nm)/NiFe (2 nm) structure the roughness effect is about 2.5 nm. We expect to have permalloy bridges through the oxide layer. The structure behaves like a granular ferromagnet above the percolation limit which is about 1.5-2 nm for permalloy layers [8]. On the other hand the films deposited by sputtering method at low Ar pressure are smooth. From Hall Effect measurements we have for sample S_3 a saturation field of about 7000 Oe. This leads to a roughness effect of about 7 nm which is very low compared with the film roughness. These electrical measurements show the importance of the deposition rate related with the Ar pressure. At low deposition rate, as we had in this work, the rate of the gaze incorporation in the layers is high and affects the conduction properties [9]. Even the sputtering method becomes in this case a not very clean one. The conduction mechanism will be mainly by multiple tunneling effects between metallic grains separated by non conducting regions due to oxides and gazes incorporated on the substrate. This effect is illustrated in Figure 6, for sample S_1. The I-V characteristic is nonlinear and confirms the presence of isolated regions between the metallic grains.

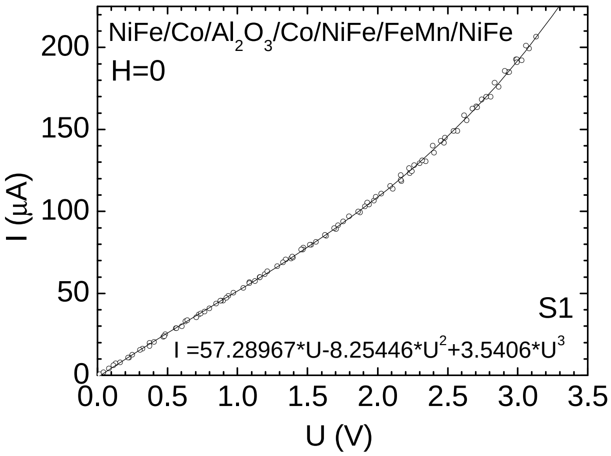

Figure 6. The conduction measurements made on the unpatterned sample, S_1.

4 CONCLUSIONS

We used electrical conductivity, tunnel effect, Hall effect and MR effect measurements to investigate the magneto-electric properties and the quality of some samples deposited by RF magnetron sputtering. At low Ar pressure we obtained structures that are rather granular films than well defined multilayer structures. Using tunneling experiments we can estimate the thickness and the quality of the oxide interlayer. By controlling the oxidation process these samples can exhibit giant Hall Effect which is very attractive for practical applications.

REFERENCES

[1] J. G. Simmons, J. Appl. Phys. **34,** 1793, 1963.

[2] T. S. Plasket, P. P. Freitas, N. P. Barradas, M. F. da Silva and J. C. Soares, J. Appl. Phys. **76,** 6104, 1994.

[3] J. S. Moodera, G. Mathon, J. Magn. Magn. Mater. **200,** 524, 1999.

[4] Y. Suezawa, Y. Gondo, J. Magn. Magn. Mater. **126**, 524, 1993.

[5] A. Gerber, A. Milner, J. Magn. Magn. Mater. **242-245**, 90, 2002.

[6] J.C. Denardin, M. Knobel, X.X. Zhang, A.B. Pakhomov, J. Magn. Magn. Mater. **262,** 15, 2003.

[7] M. Volmer, J. Neamtu, J. Magn. Magn. Mater. **272-276**, 1881, 2004.

[8] T. Lucinski, G. Reiss, N. Mattern, L. van Loyen, J. Magn. Magn. Mater. **189,** 39, 1998.

[9] Milton Ohring, The Materials Science of Thin Films, Academic Press, 95-96, 1992, ISBN 0-12-524990-X